

TRƯỜNG ĐẠI HỌC QUY NHƠN
QUY NHON UNIVERSITY

TẠP CHÍ KHOA HỌC
TRƯỜNG ĐẠI HỌC QUY NHƠN

QUY NHON UNIVERSITY
JOURNAL OF SCIENCE

KHOA HỌC TỰ NHIÊN VÀ KỸ THUẬT
NATURAL SCIENCES AND ENGINEERING

19 (5)

2025

OCTOBER 2025

CONTENTS

1.	Fabrications of chitosan-derivative-based superabsorbent materials for agriculture application Cao Luu Ngoc Hanh, Nguyen Tuong Vy, Huynh Lien Huong, Nguyen Minh Nhut, Pham Mai Huong	5
2.	GAAFETs and 2D materials for sub-3 nm nodes: performance, fabrication challenges, and integration strategies Huynh Cong Tu, Hoang Anh Phuong	25
3.	ZIF-67-derived Co ₃ O ₄ porous crystalline material for electrochemical ascorbic acid detection Ngo Thi Thanh Hien, Nguyen Vu Ngoc Mai	43
4.	Fabrication of ovalbumin-loaded alginate-chitosan nanoparticle using coaxial electrospray for protein delivery orientation Cao Luu Ngoc Hanh, Nguyen Tuong Vy, Pham Duy Toan, Nguyen Thi Bich Thuyen, Dang Huynh Giao, Le Truong Thinh	53
5.	Recurrent neural network models for wind power forecasting Van Ninh Nguyen, Xuan Tham Nguyen, Van Thong Than, Thanh Bao Doan, Ngoc Toan Luong, Tuan Ho Le	69
6.	Study on the effect of pure fermentation starters (banh men) on the quality of fermented glutinous rice (com ruou) Le Bich Tuyen, Huynh Van Quoc Canh, Truong Thi Tu Tran	81
7.	A characterization of Zygmund-type spaces and its application Thai Thuan Quang, Nguyen Van Dai	93
8.	Study on optimal acceleration control of three phase induction motors Do Van Can, Than Van Thong, Hoang Van Hieu	103
9.	The free energy function of Ising model in a strong magnetic field Nguyen Tong Xuan	117
10.	Design of high gain, low side-lobe level base station array antenna for 5g-sub 6 ghz communication system Dang Thi Tu My, Ho Van Phi, Huynh Nguyen Bao Phuong	129
11.	Synthesis of bioplastic films from <i>Vitex negundo</i> L. leaf extract based on sweet potato starch and investigation of antibacterial activity of the products Pham Khanh Nguyen Huan, Le Hong Ngoc Bao, Nguyen Thi Kieu Tien, Ha Thi Kim Quy	139

Chế tạo vật liệu siêu hấp thụ dựa trên dẫn xuất chitosan ứng dụng trong nông nghiệp

Cao Lưu Ngọc Hạnh^{1,2,*}, Nguyễn Tường Vy², Huỳnh Liên Hương¹,
Nguyễn Minh Nhật¹, Phạm Mai Hương¹

¹Khoa Kỹ thuật Hóa học, Trường Đại học Cần Thơ, Việt Nam

²Phòng thí nghiệm vật liệu composite, Khoa Kỹ thuật Hóa học, Trường Đại học Cần Thơ, Việt Nam

Ngày nhận bài: 28/02/2025; Ngày sửa bài: 09/06/2025;

Ngày nhận đăng: 25/06/2025; Ngày xuất bản: 28/10/2025

TÓM TẮT

Nghiên cứu đã tận dụng thành công vỏ tôm để chiết tách chitosan bằng quy trình deacetyl hóa hai bước với độ deacetyl hóa cao (91,48%). Hơn nữa, chitosan được gắn các nhóm chức ưa nước để tạo thành O-carboxymethyl chitosan (O-CMCS), nhằm tăng độ tan của vật liệu trong môi trường trung tính và kiềm. Tiếp theo là ghép dẫn xuất chitosan với axit acrylic (AA) để tạo ra vật liệu polyme siêu hấp thụ từ O-CMCS (O-CMCS-SAP). Các thông số tối ưu để tổng hợp vật liệu là nồng độ kali persulfate 2%, tỷ lệ O-CMCS:AA là 1:8 và nồng độ glutaraldehyde 2%, tạo ra vật liệu SAP với khả năng hấp phụ tối đa là 658,16 g/g sau 15 ngày ngâm trong nước. Khả năng phân hủy sinh học sau bốn tuần thử nghiệm và giữ nước để cung cấp cho cây trong quá trình sinh trưởng là điểm mạnh của vật liệu O-CMCS-SAP. Ngoài ra, nghiên cứu đã thành công trong việc phân tích sự hình thành và sinh trưởng của hai cây ớt giống nhau, nhưng khác nhau ở việc có và không có sử dụng O-CMCS-SAP. Cây ớt sử dụng O-CMCS-SAP có khả năng sinh trưởng và phát triển cao hơn nhiều so với cây không sử dụng O-CMCS-SAP. Sau tám tuần thử nghiệm, những cây không được tưới nước cho thấy lá héo và phát triển cực kỳ thấp. Dựa trên tất cả các phát hiện thực nghiệm, vật liệu O-CMCS-SAP đã được tổng hợp thành công và có tiềm năng được ứng dụng để cải tạo đất khô cằn.

Từ khóa: Vật liệu siêu hấp thụ, dẫn xuất chitosan, trương nở, giữ nước.

*Tác giả liên hệ chính.

Email: clnhanh@ctu.edu.vn

Fabrications of chitosan-derivative-based superabsorbent materials for agriculture application

Cao Luu Ngoc Hanh^{1,2,*}, Nguyen Tuong Vy², Huynh Lien Huong¹,
Nguyen Minh Nhut¹, Pham Mai Huong¹

¹Faculty of Chemical Engineering, Can Tho University, Vietnam

²Composite Materials Lab, Faculty of Chemical Engineering, Can Tho University, Vietnam

Received: 28/02/2025; Revised: 09/06/2025;

Accepted: 25/06/2025; Published: 28/10/2025

ABSTRACT

The study successfully utilized shrimp shells to extract chitosan using a two-step deacetylation process with a high degree of deacetylation (91.48%). Furthermore, chitosan is attached with hydrophilic functional groups to form O-carboxymethyl chitosan (O-CMCS), to increase the solubility of the material in neutral and alkaline environments. The chitosan derivative was grafted with acrylic acid (AA) to produce a superabsorbent polymer (SAP) material from O-CMCS (O-CMCS-SAP). The ideal conditions for the material's synthesis were a 2% glutaraldehyde concentration, a 1:8 O-CMCS:AA ratio, and a 2% potassium persulfate concentration that created SAP material had a maximum adsorption capacity of 658.16 g/g after a 15-day immersion in water. The ability to biodegrade after four weeks of testing and retain water to provide plants during growth were the strengths of the O-CMCS-SAP material. Furthermore, the study effectively examined the same two chili plants developed and changed in the presence and absence of O-CMCS-SAP. Compared to plants lacking O-CMCS-SAP, the chili plants with O-CMCS-SAP grew and developed significantly more. Following eight weeks of testing, the water-deficient plants had wilting leaves and remarkably slow development. The O-CMCS-SAP material was effectively synthesized and may be used to enhance arid soil, according to all experimental results.

Keywords: Superabsorbent materials, chitosan derivatives, swelling, water retention.

INTRODUCTION

Superabsorbent polymers (SAP) are a kind of three-dimensional crosslinked hydrophilic polymers, which can absorb and retain large amounts of water up to several hundred times their own weight.¹ Because of their excellent properties, SAP are widely used in various fields, such as hygiene product, agriculture, construction industry, petroleum industry, adsorption and materials for controlled release devices.² SAP are currently the subject of extensive

research in Vietnam and are gradually being commercialized. However, modified starch - which may compete with human and animal food sources - is the primary basic material used by the majority of authors. Khoi et al. successfully created the SAP for soil (AMS-1). The product known as AMS-1 is made by grafting acrylic acid into cassava starch, which has a unique capacity to store and absorb water. AMS-1 is a fine, ivory-white powder that will swell 400 times its actual weight when exposed

*Corresponding author.

Email: clnhanh@ctu.edu.vn

to water. It can be stored in soil for 10 to 12 months. AMS-1 has a wide range of uses in agricultural production, including helping to improve the physical and chemical properties of soil and acting as a moisture-retaining agent on hillsides where water readily drains away or on dry, sandy soils. Apart from its ability to absorb water, AMS-1 can also absorb other solutions, such as saline and urine.³

Furthermore, Lac Trung Company produced the superior product known as superabsorbent polymer (SAM-1) in 2006. The product is the end product of the polymerization of modified cassava starch and acrylic acid. Each tiny moisture-retaining particle can absorb up to 350–400 g/g of water and then gradually release it to provide moisture for the plant during times when there is no rain or irrigation water. The primary ingredient is cassava starch, and chemical technology is used to create a type of polymer that has the ability to absorb and retain water. Watering will be much decreased after 30 to 45 days when the particles will release all of the water, the moisture-retaining particles will revert to their initial size, and they will continue to collect water when it rains or is watered. Moisture-retaining particles should be used for six to eighteen months before being replaced. Utilizing the product has been shown to reduce plant mortality by over 100%, lower plant care expenses by up to 80%, transport crops across great distances, and harvest crops 20% earlier than planned.⁴

Sutradhar et al. synthesized superabsorbent polymers in 2015 by combining gamma radiation with carboxymethylated cellulose (CMC) and acrylic acid (AA). In particular, the author synthesized by copolymerizing carboxymethylated cellulose and acrylic acid and then irradiating it with Co-60. Due to the extremely high energy of the gamma irradiation source, initiators, catalysts, and cross-linking agents are not required in the synthesis technique. At the same time, a radiation dose of 5 kGy was shown to have the maximum adsorption

capability. According to experimental results, the product's high water retention capacity had a favorable impact on the germination of okra and wheat seeds as well as the growth of young plants. Thus, it can be said that CMC/AA, which has a lot of great qualities, can be employed as a regulator in agriculture, particularly in desert and arid areas.⁵

A superabsorbent water gel (CHCAUR) was created by Narayanan et al. using urea, citric acid, and chitosan. A superabsorbent material was developed from renewable resources such as chitosan, citric acid and urea by hydrothermal method. The maximum adsorption was about 1250 g/g in distilled water and 210 g/g in 0.1% sodium chloride solution. It is demonstrated that the water absorption mechanism results from the electrostatic attraction of water for ionic bonding, the presence of pores, and the undulating surface caused by the development of nanofibers. It was demonstrated that physical bonds, such as electrochemical bonds, macropores, and morphological characteristics, are the source of the water absorption. It was found that CHCAUR had a very high chitosan content and a low urea/citric acid content, making it highly biodegradable in soil. With a significant and regulated water output, the product was employed to supply nutrients to the soil. CHCAUR was also shown to dramatically lower the rate of water evaporation when applied to soil.⁶

In 2018, Neamjan et al. reported on the biodegradable superabsorbent polymer based on cellulose from sugarcane bagasse (SCB) for water resource management, horticulture and other products in agricultural applications. Specifically, the superabsorbent polymer was prepared by polymerizing SCB monomer and AA in the presence of N, N-methylene-bisacrylamide as a cross-linker using twin screw extruder and using ammonium persulfate/sodium sulfite (APS/SS) as a pair initiator in an enhanced redox system because SS is a reducing agent that can enhance the polymerization reaction. The

results indicated that the soil moisture retention capacity could be enhanced by applying SCB superabsorbent polymer. Therefore, the superabsorbent polymers prepared in this study have the potential to be used as a water source in agriculture.⁷

According to Yassine et al., the extracted cellulose in superabsorbent materials with high water absorption capabilities was oxidized by sodium periodate in a controlled manner to produce cellulose dialdehyde with a regulated aldehyde content. Using N,N-methylene-bis-acrylamide as a cross-linking agent and potassium persulfate as an initiator, the cellulosic materials were mixed with a suspension of acrylic and itaconic acids to create composite hybrid hydrogels through radical chain polymerization in water. Notably, dialdehyde cellulose has the potential to improve the hydrogel's swelling and water retention ability because of its strong polarity, low crystallinity index, and hydrophilicity. With a dialdehyde cellulose content of 5%wt, synthesized materials demonstrated a water absorption capacity of up to $1240 \pm 60 \text{ g.g}^{-1}$.⁸

One of the most important issues of concern today in the agricultural sector is watering plants during the drought season while still ensuring that enough water is provided for the plants during their growth and development, avoiding waterlogging due to over-watering or reducing the mortality rate of plants during the dry season, etc. To help solve this problem, a new material has been researched and widely applied by many researchers in many places around the world, which is superabsorbent polymer (SAP). This material is widely used in agriculture, sanitary napkins, medicine, and other fields because it can absorb and retain a lot of water in its structure - many times more than its dry weight. In the agricultural sector, SAP application can lower plant mortality rates from dehydration, reduce care, watering frequency, water amount, pesticide residues, pre-season harvesting, etc. Nonetheless, modified starch and acrylic acid

provide the majority of the produced SAP.^{9,10} However, the use of starch to synthesize SAP results in a very serious waste of food sources, a source of nutrition for both humans and animals, as well as intense competition between humans and animals for food sources. This is because starch is still an important food source, both for humans and animals. Therefore, in this study, we extract chitosan for SAP manufacture utilizing shrimp shells for by-product utilization, instead of starch.¹¹ The primary drawback of CS is its poor solubility in neutral and alkaline pH environments. For this reason, in this study, we modify CS and produce CS derivatives (by adding hydrophilic groups to the CS structure to make CS more soluble in an environment with a pH greater than 6.5) in order to synthesize SAP. Since SAP materials are made from natural raw materials, such as seafood waste, they are highly biodegradable, environmentally benign more than SAP materials derived from synthesis polymer like polyacrylic acid and polyvinyl alcohol,^{12,13} and satisfy the "green chemistry" standards that our nation is striving for in the future while creating new materials. Research on the synthesis of biodegradable superabsorbent polymer (SAP) from chitosan derivatives and its application in agriculture based on the following steps: synthesis of SAP material, analysis of adsorption-influencing factors, analysis of adsorption and desorption capacity, and analysis of the product's biodegradability over time in a soil environment.

2. MATERIALS AND METHODS

2.1. Materials

Fresh shrimp shells are gathered from the Chau Thanh District of Kien Giang Province's Bright Star Seafood Company Limited. Following gathering, the shrimp shells undergo numerous water washes to clean them before being dried and used.

Sodium hydroxide (NaOH, 99%), oxalic acid ($\text{C}_2\text{H}_2\text{O}_4$, 99.6%), hydrochloric acid (HCl), potassium persulfate ($\text{K}_2\text{S}_2\text{O}_8$), acetic

acid (CH_3COOH , 99.5%), chloroacetic acid (ClCH_2COOH), acrylic acid ($\text{CH}_2=\text{CHCOOH}$, 99.5%), and glutaraldehyde ($\text{C}_5\text{H}_8\text{O}_2$, 50%), KMnO_4 , 2-propanol (99.5%), and ethanol ($\text{C}_2\text{H}_5\text{OH}$, 99%) were acquired from Xilong, China. All other chemicals were of reagent grades or higher and used as received.

2.2. Preparation of chitosan derivatives from shrimp shells by chemical method

The chemical process of creating chitosan derivatives from shrimp shells was carried

out in two stages. Chitosan was first extracted from shrimp shells using a chemical process that involved two rounds of deacetylation. This was followed by the production of chitosan derivatives using O-carboxymethyl. The synthesis of chitosan from shrimp shells was based on the optimal parameters of the synthesis process of Kurniasih et al. and several steps were modified to fit Vietnamese research settings.¹⁴ The synthesis diagram is presented specifically in Figure 1.



Figure 1. Chitosan preparation using shrimp shells.

The shrimp shells were initially collected at Seafood Company, the shrimp shells were refrigerated with ice while being transported. After processing, the shrimp shells were cleaned to get rid of any remaining shrimp meat, debris, and contaminants. To preserve the shrimp shells for the upcoming tests, they were then dried. Following first treatment, the shrimp shells were soaked in a 2M NaOH solution for two days in order to separate the protein and fat. At this point, alkali at pH 11–12 and room temperature degraded the protein to produce soluble free amines that were simple to remove after the shrimp shells were cleaned with water. Following multiple water washes, the product was ultimately cleaned with distilled water until its neutral pH reached. The shrimp shells were then demineralized using a 1M HCl solution for 48 hours at room temperature in order to extract

calcium and a few other minerals. Crude chitin was treated with 1% oxalic acid and 1% KMnO_4 oxidant for 15 minutes at room temperature in order to eliminate the pale pink astaxanthin color. Lastly, a 48% NaOH solution was used to deacetylate crude chitin ($-\text{NHCOCH}_3$) to chitosan ($-\text{NH}_2$) for 48 hours at room temperature. To produce chitosan with a larger degree of deacetylation, the deacetylation step was carried out again, but at a higher temperature (80°C). Chitosan was dried for 12 hours at 50°C in order to preserve it.

The degree of deacetylation of the obtained chitosan was determined by acid-base titration method.¹⁵ A 0.1 g crude chitosan sample was dissolved in 30 mL of 0.1 M HCl at room temperature, and 5–6 drops of methyl red were added. The solution was then titrated with 0.1 M NaOH, and the titration was stopped when

the solution turned orange. The DDA of crude chitosan was calculated based on the number of mL of 0.1 M NaOH used during the titration. The degree of deacetylation was calculated according to the formula (1):

$$\text{Deacetylation (\%)} = \frac{(C_1 V_1 - C_2 V_2)}{M \times 0.0994} \times 100 \quad (1)$$

Where C_1 was concentration of HCl solution used to dissolve chitosan (M), C_2 was concentration of NaOH solution used for titration (M), V_1 was volume of HCl solution (mL), V_2 was volume of NaOH solution used in titration (mL), M was mass of crude chitosan sample (g).

The method of Mohamed et al. provides the basis for the synthesis of O-carboxymethyl chitosan (O-CMCS),¹⁶ which has high solubility in neutral and alkaline pH by attaching hydroxyl groups to the chitosan structure.

However, certain variables have been modified to suit Vietnam. Figure 2 specifically displays the O-CMCS synthesis diagram. First, in a three-necked flask, 100 mL of 20% NaOH solution was mixed with 10 g of second deacetylated chitosan (CS-DD2) and stirred for 15 minutes. The carboxymethylation process was then carried out by adding 15 g of monochloroacetic acid and 20 mL of 2-propanol dropwise to the alkalized liquid and keeping the reaction at 40°C for two hours. 10% acetic acid was used to neutralize the liquid after two hours, bringing its pH down to 7. After removing insoluble substances with 70% EtOH, the product was washed with absolute EtOH. Finally, the O-CMCS product was then dried at 50°C. Environmentally harmful chemicals used in the extraction and modification of chitosan such as strong base and acid are reused and then neutralized before disposal.

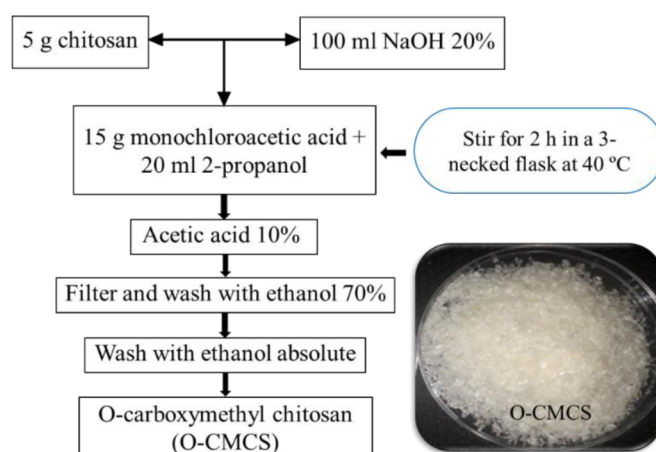


Figure 2. Synthesis diagram of O-CMCS synthesis from chitosan.

2.3. Synthesis of O-CMCS-SAP material from O-CMCS

Superabsorbent polymer (SAP) was synthesized from chitosan derivative (O-CMCS) in a 250 mL glass beaker that was submerged in silicone oil and had a magnetic stirrer that was heated to 60 - 70 °C throughout the reaction. The experiment was carried out by dispersing 1 g of O-carboxymethyl chitosan (O-CMCS), which was synthesized in the previous step, evenly in distilled water. Next, the initiator ($K_2S_2O_8$), monomer ($CH_2CHCOOH$), and cross-linker

(glutaraldehyde) were added one after the other while being continuously stirred on a heated magnetic stirrer for a predetermined amount of time. The reaction mixture from the previous step was then allowed to cool to room temperature and neutralized with 3% NaOH to reach neutral pH. Methanol was then added while stirring. Following the completion of the reaction, the product is crushed and dried for 48 hours at 50°C. At this point, O-CMCS-SAP material is obtained. In particular, Figure 3 displays the material synthesis diagram.

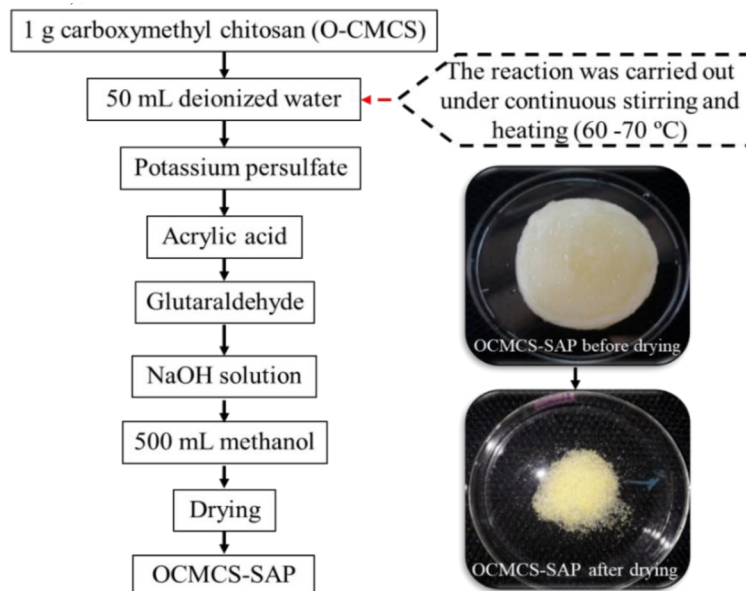


Figure 3. Synthesis diagram of O-CMCS-SAP.

2.4. Characterizations

The surface images of the samples, including first deacetylated chitosan, second deacetylated chitosan, O-carboxymethyl chitosan, and superabsorbent materials from O-carboxymethyl chitosan, were observed using an optical microscope (Nikon EPIPHOT 200, Japan) and a scanning electron microscope (SEM, S-4800, Hitachi, Japan). The chemical structure of the investigated samples was investigated using Fourier-transform infrared spectroscopy (FTIR, Nicolet 6700, Thermo Scientific, USA). The scanning was performed 32 times under the condition of the wave number of 4000-400 cm^{-1} and the resolution of 4 cm^{-1} . The crystallographic structure was determined using X-ray diffraction (XRD, Empyrean, PANalytical). The material's thermal stability was assessed using thermogravimetric analysis (TGA, Q500 V20.10), which determined the material's temperature range of 50 to 600°C in an inert nitrogen environment at a rate of 5°C per minute.

2.5. Evaluation of water absorption and desorption

The water absorption ability of O-CMCS-SAP was evaluated as previous study, with a

minor modification. A certain amount of SAP material was dried to consistent mass and then immersed in distilled water at room temperature. Everyday within first 30 days, the samples were taken and removed water on the surface for absorption quantitation, the absorbency (g/g) of material was calculated by applying formula (2), considering the mass variation before and after immersion in water:

$$\text{Absorbency (g/g)} = \frac{m_{\text{swollen}} - m_{\text{initial}}}{m_{\text{initial}}} \quad (2)$$

where m_{initial} and m_{swollen} are the weights of SAP material in initial and swollen states, respectively.

The O-CMCS-SAP material in the maximum water absorption state was investigated desorption property which occurred within 30 days, and the implementation process that was similar to adsorption section. The desorption percentage was computed as the following formula (3):

$$\text{Desorption (\%)} = \frac{m_{\text{max}} - m_{\text{day}}}{m_{\text{max}}} \times 100 \quad (3)$$

where m_{max} and m_{day} are the weights of SAP material in initial and swollen states, respectively.

2.6. Biodegradation study by soil-burial method

To examine the biodegradability of O-CMCS-SAP material, soil burial test was carried out on

a laboratory scale. The dried and swollen states of samples were buried in the soil (included arid soil and 28% humid soil) and thus subjected to the action of microorganisms. Everyweek, the samples were taken out and the soil on the surface is removed to determine the mass. The weight loss of the samples indicates the amount of biodegradation in soil environment.

2.7. Application of O-CMCS-SAP material on agriculture

To demonstrate the applicability of O-CMCS-SAP material in agriculture, they were tested on commercial chili plants. Two chili plants were prepared, then a certain amount of SAP material was added to the soil of one plant by burying it near the roots of the plant, and the soil of the other plant was not mixed with SAP material and used for comparison. They were monitored for growth by observing their morphology.

3. RESULTS AND DISCUSSION

The chitosan samples that were deacetylated once (CS-DD1) and twice (CS-DD2) had clear color variations, as seen in Figures 4a and 4b. The CS-DD2 product was bright white, whereas the CS-DD1 product was ivory yellow. Both products shared the trait of being odorless. Through the acid-base titration method, the deacetylation degree of CS-DD1 sample was 79.68% and the deacetylation degree of CS-DD2 sample was 91.48%, which were much higher than those of CS-DD1 sample. The extraction process with two deacetylation steps achieved an overall extraction efficiency of chitosan from shrimp shells of 53.28%.



Figure 4. CS-DD1 (a) and CS-DD2 (b).

3.1. Investigation results of factors affecting the adsorption capacity of O-CMCS-SAP

3.1.1. Effect of potassium persulfate (KPS) concentration

By adjusting the KPS concentration from 1 to 2.5% while maintaining the same reaction temperature (60 - 70°C), O-CMCS:AA ratio (1:6), and glutaraldehyde concentration of 1.5%, the impact of KPS concentration on water adsorption was examined. In Figure 5, the water adsorption outcomes of four samples with varying KPS concentrations are explicitly displayed.

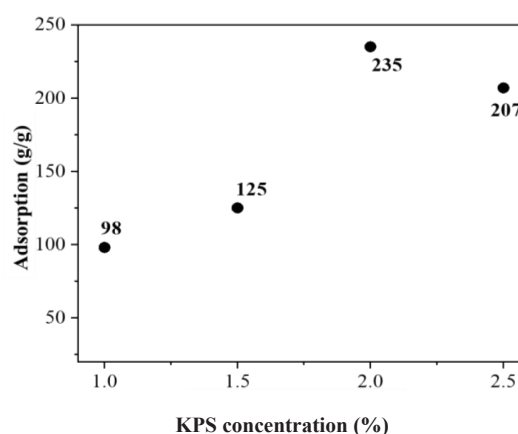


Figure 5. Results of the influence of KPS concentration on water adsorption of O-CMCS-SAP.

The impact of the initiator (KPS) on the polymer's swelling was depicted in Figure 5. As the KPS concentration rose from 1 to 2.5%, the swelling ratio of the polymer in distilled water changed. This was because the higher KPS concentration (1 - 2%) led to a greater number of radical formation, and more polymer chains were created that was efficient in holding water molecules resulted in higher water adsorption. As Figure 5 illustrates, a KPS concentration of 2% was appropriate for the development of extremely inflated products. The material's adsorption capacity in distilled water, however, decreased as the KPS concentration was raised further because more free radicals would promote the reaction termination step through the combination of free radicals. Furthermore, the shorter kinetic chain length due to excessive initiation causing the polymer network to

become loose, less cross-linked and less able to absorb water.

3.1.2. The effect of O-CMCS:AA ratio

The results of the water adsorption of four samples are shown in Figure 6 specifically. The effect of acid acrylic (AA) on water adsorption was investigated by changing the O-CMCS:AA ratio from 1:2 to 1:10 while maintaining the parameters such as reaction temperature 60 - 70°C, KPS concentration of 2%, and glutaraldehyde concentration of 1.5%. Since AA is the primary water adsorption group, more AA molecules will be grafted into the O-CMCS structure as the AA ratio rises. This will boost the material's water adsorption and make O-CMCS-SAP more hydrophilic. However, increasing the O-CMCS-SAP:AA ratio to 1:10 only increases the material's water adsorption by 5% (insignificant), hence the O-CMCS-SAP ratio of 1:8 is the best option for creating a material with high water adsorption while remaining economical. On the other hand, increasing the AA concentration too high reduces the material's swelling ratio because increasing the AA concentration raises the viscosity of the product and inhibits the mobility of free radicals, lowering the ability to absorb water.^{1,2,17} Furthermore, excess AA that was not fully incorporated into the O-CMC network could polymerize independently, forming separate poly(acrylic acid) segments or inert gels without superabsorbent function.

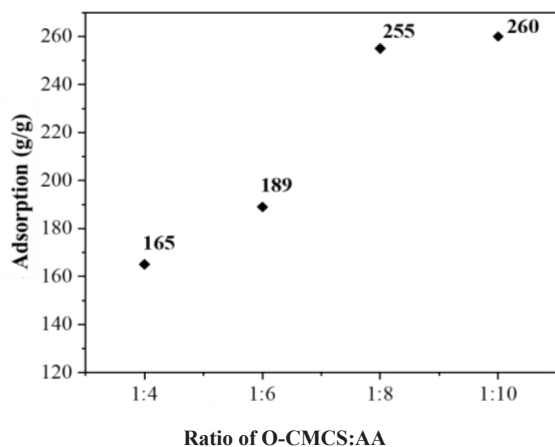


Figure 6. Results on the effect of AA concentration on water adsorption.

3.1.3. Effects of glutaraldehyde concentration

The effect of glutaraldehyde concentration on water adsorption was investigated by increasing the glutaraldehyde concentration from 1 to 3% while maintaining the parameters such as reaction temperature 60 - 70°C, KPS concentration 2%, and O-CMCS:AA ratio 1:8. The water adsorption results of 5 samples are shown specifically in Figure 7. When the amount of cross-linking agent (glutaraldehyde) is insufficient, the swelling ratio of O-CMCS-SAP is low, as the low cross-linking density cannot sustain the adsorbed water. Simultaneously, the material's capacity to adsorb water steadily increases as the amount of glutaraldehyde grows due to the flexible network creation facilitated good water retention, and the swelling ratio achieves its maximum value when the glutaraldehyde concentration approaches 2%. Because excessive cross-linking produces a dense network structure that results in poor water adsorption, the material's absorption capacity falls as cross-linking increases. This indicates that 2% glutaraldehyde is appropriate for producing a product with a high water absorption rate.

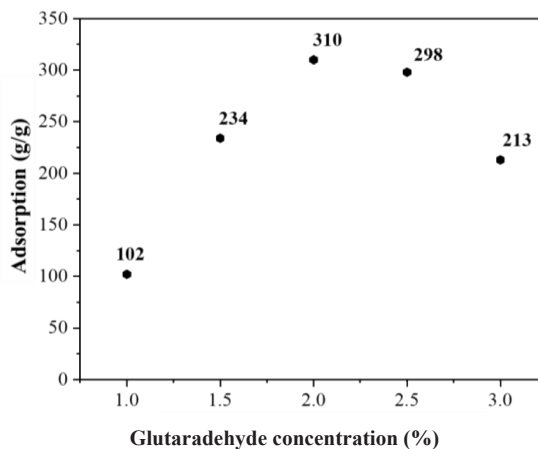


Figure 7. Results on the effect of glutaraldehyde concentration on water adsorption.

3.2. Investigation of adsorption and desorption capacity of O-CMCS-SAP materials

The investigation into the water absorption of O-CMCS-SAP material lasted for 30 days at room temperature (~30°C) and the material was utilized in powder form. The findings of the

material's water absorption ability are shown in particular in Figure 8.

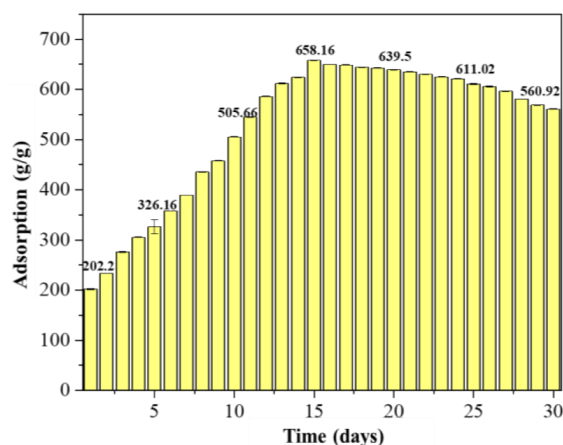


Figure 8. Results on water absorption capacity of O-CMCS-SAP material in 30 days.

After five days, the material's water absorption capacity grew steadily from 202.2 g/g to 326.16 g/g, and after ten days, it reached 505.66 g/g. Nevertheless, the material's capacity to absorb water rose further, reaching a very high 658.16 g/g when the duration was extended to 15 days. This is because, as survey time increases, so do the hydrogen bonding and Van der Waals (collision) forces between water molecules and the super adsorbent. The water absorption trend, however, declined dramatically after 15 days, as evidenced by the material's water absorption capacity dropping to 639.5 g/g on day 20, continuing to decline accordingly on day 25, and reaching only roughly 560.92 g/g on day 30. The primary cause of inadequate water absorption is the material's degradation during 15 days of swelling, which is regarded as the adsorption release period if it lasts for 15 days or longer and occurs after the material has attained the ideal adsorption condition.

Following the examination of the material's water adsorption capacity, a 30-day test of the material's water desorption capability was also conducted, as shown in Figure 9. In particular, the material only discharged only 2.4% of its water content in the first five days, but over time, this percentage grew steadily. The material's

adsorption release trend significantly increased starting on day 15. The material usually released approximately 85.5% on day 16, and then started to leak substantially (~78%) on day 17. This is because evaporation on the material's surface over time and osmotic pressure differ. The material saw a strong adsorption release phase from day 25 to day 30, and as the graph illustrates, after 30 days, the material achieved an adsorption release of roughly 49.5% (relatively lower than the initial weight).

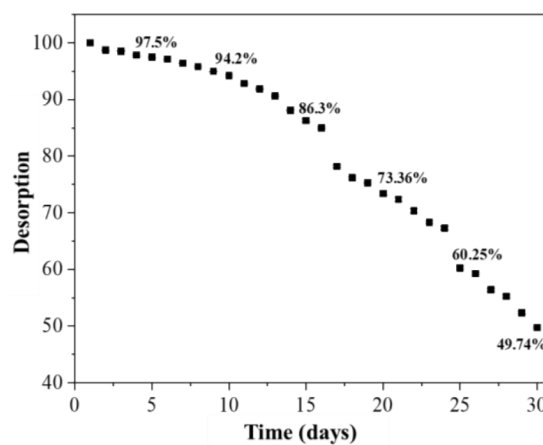


Figure 9. Material desorption test graph after 30 days.

3.3. Physicochemical characteristics

The characteristics of the shape and surface structure of CS-DD1, CS-DD2, O-CMCS, and O-CMCS-SAP samples were observed with an optical microscope at 50x magnification, shown in Figure 10. Because the fibers contracted, the surface morphology of CS-DD1 showed numerous pores in Figure 10a; however, because these pores were rather narrow, the material was unable to achieve its super water adsorption objective. Following the second DD with 48% NaOH (80 °C), the CS-DD2 sample's surface morphology is shown in Figure 10b. The initial fibers started to straighten and were easily visible when the surface of the CS-DD2 sample displayed an image of long, straight fibers. The surface morphology of the CS derivative (O-CMCS) is vividly displayed in Figure 10c. In comparison to the CS-DD2 sample, the O-CMCS surface no longer contains dense fibers like CS-DD2,

but instead has sparser fibers. Additionally, the network connection has expanded, making it easier for water to enter the structure. Because the original structure of CS was destroyed when it was modified with monochloroacetic acid (MA), the network structure expanded when the fibers became sparser. These expanded network holes are crucial for the adsorption of water into the polymer structure, demonstrating that O-CMCS is a potential source of raw materials for the synthesis of SAP.

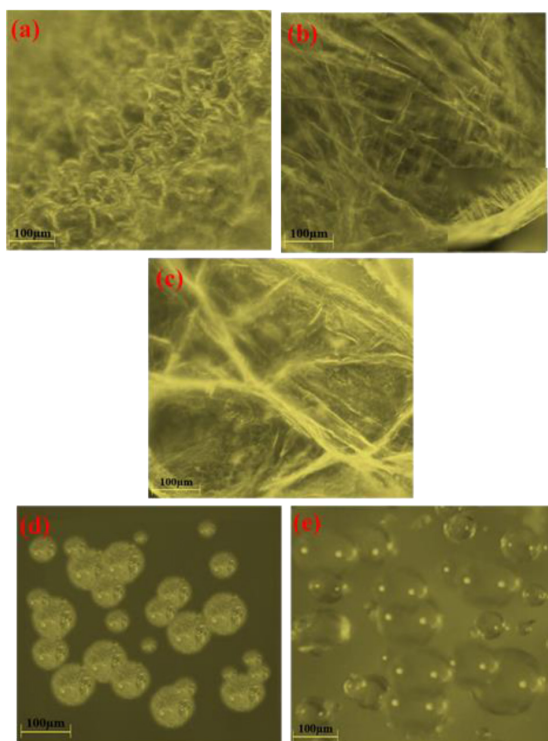


Figure 10. Optical microscope images of CS-DD1 (a), CS-DD2 (b), O-CMCS (c), O-CMCS-SAP (d), and OCMCS-SAP after 3 days of swelling (e).

The O-CMCS-SAP material's morphology before and after swelling is depicted in Figures 10d and 10e. Despite their spherical form and relatively modest size ($d_1 = 96 \pm 0.06 \mu\text{m}$), the resulting O-CMCS-SAP particles exhibit polydispersity, as seen in Figure 10d. But after swelling for three days ($d_2 = 373 \pm 0.03 \mu\text{m}$), the O-CMCS-SAP particles in Figure 10e had a diameter that is four times greater than the original particles. After reaching the adsorption state (276.5 g/g), the material particles

swelled; however, this is not the material's ideal adsorption state, thus the microparticles will keep growing until the material reaches the ideal adsorption state (15 days of swelling). Optical microscope results showed that the network structure expanded with each stage of production, resulting in a more hydrophilic material. The substance usually had a lot of water after three days of swelling, which resulted in the formation of spherical microparticles. These microparticles are in charge of giving the plant water as it grows and develops.

A scanning electron microscope was then used to examine the OCMCS-SAP material's shape and surface structure both before and after swelling (Figure 11). The OCMCS-SAP particles' surface first developed pores, but in certain places, agglomeration was noticed as a result of overheating during the drying process. After three days of swelling, the material's surface (Figure 11b) changed morphologically as the original pores progressively grew larger. The surface morphology of the material seen was more distinct than in the optical microscopy image, and the presence of pores in the product following adsorption showed that the synthesized material particles were successful and that these pores were crucial in absorbing water to supply water for plants.

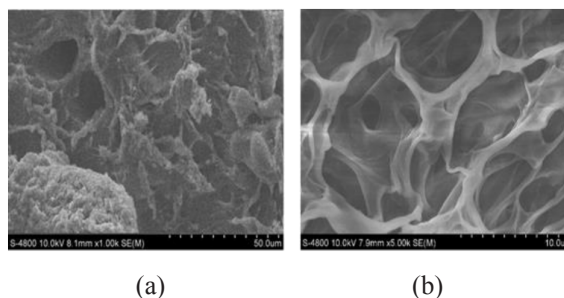


Figure 11. SEM images of (a) O-CMCS-SAP and (b) OCMCS-SAP after 3 days of swelling.

Figure 12 illustrates the differences in the functional groups of the newly synthesized materials based on the infrared spectra of the CS, O-CMCS, and O-CMCS-SAP samples after they were dried.

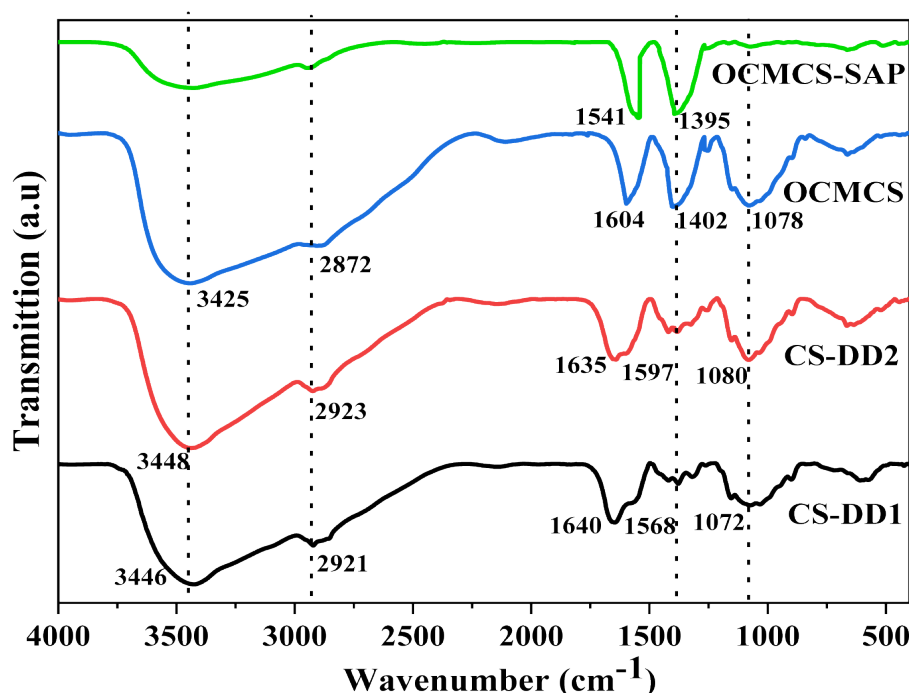


Figure 12. FTIR spectra of CS-DD1, CS-DD2, O-CMCS, and O-CMCS-SAP samples.

Characteristic vibrations in the FTIR spectrum of CS include O-H and N-H stretching vibrations in the 3350-3450 cm^{-1} range and deformation vibrations of the fatty chain C-H bonds in the 2920-2860 cm^{-1} , 1420 cm^{-1} , and 1380 cm^{-1} ranges, respectively. In-plane N-H bending vibrations are responsible for the formation of high intensity peaks at 1640 cm^{-1} on CS-DD1 and 1635 cm^{-1} on CS-DD2, which is the distinctive peak of polysaccharide on CS. The ether and cyclic glycoside linkages in the CS structure are simultaneously visible in the absorption in the 1000-1300 cm^{-1} range.¹⁸

The FTIR spectrum of O-CMCS shows the appearance of new peaks with high intensity at wave numbers 1604 cm^{-1} and 1402 cm^{-1} , these bands appear corresponding to the stretching vibration of the C=O bond and the $-\text{CH}_2\text{COOH}$ vibration, through which we can confirm the appearance of the carboxymethyl group. In addition, the peak 1380 cm^{-1} no longer appears in the spectrum of the CS derivative, which proves that the C-H bond was partially removed when CS reacted with MA.¹⁹

An additional carboxymethyl chitosan main chain is present in the superabsorbent polymer product's structure, as indicated by the peak in the 1541 cm^{-1} region of the O-CMCS-SAP FTIR spectrum. When it appears at the peak of 1395 cm^{-1} , this distinctive band, which is associated with the asymmetric stretching of the carboxylate anion group, is likewise extremely intense. Additionally, there are no odd peaks in the superabsorbent material's FTIR spectra. The O-CMCS-SAP product was successfully created when the polymer's AA was grafted onto O-CMCS when glutaraldehyde was present, according to the FTIR data.¹⁷

X-ray diffraction was used to examine the XRD structures of CS-DD2, CS derivatives, and superadsorbed products (Figure 13). Three primary strong diffraction peaks at $2\theta=10.1^\circ$, 20.02° , and 22° with high intensity were visible in the XRD analysis results of CS, as illustrated in Figure 8, suggesting that the produced CS had high crystallinity.²⁰ The production of many hydrogen bonds in the crystal lattice, which resulted in high CS hardness and the development of crystalline areas, was the

cause of the high crystallinity of CS. However, following chemical modification, the diffraction peak of the CS derivative (O-CMCS) displayed a decrease in the characteristic peak. This was because the carboxymethyl groups were introduced, increasing the distance between molecular chains and destroying the original structure. Additionally, the number of hydrogen bonds decreased, resulting in an amorphous structure of O-CMCS.²¹ The amount of acrylic acid grafted on the CS derivative structure results in a decrease in crystallinity, which is especially noticeable in the XRD pattern of the superabsorbent product (O-CMCS-SAP) when compared to the CS and O-CMCS samples. According to the three materials' XRD data, CS has the highest crystallinity, as seen by

the distinctive, highly intense peaks on CS. O-CMCS, which has monochloroacetic acid (MA) grafted onto it, has a lesser crystallinity following chemical transformation. Last but not least, the O-CMCS-SAP material sample exhibits the lowest crystallinity,²² demonstrating the amorphous structure of the composite material and its high water adsorption capacity as a result of its pores. The success of the synthesized material was demonstrated by the XRD analysis of three samples, CS-DD2, O-CMCS, and O-CMCS-SAP, as shown in Figure 13. The appearance of distinctive peaks in CS and the decrease in crystallinity following chemical transformation, which results in the material having an amorphous structure, demonstrate the success of the synthesis.

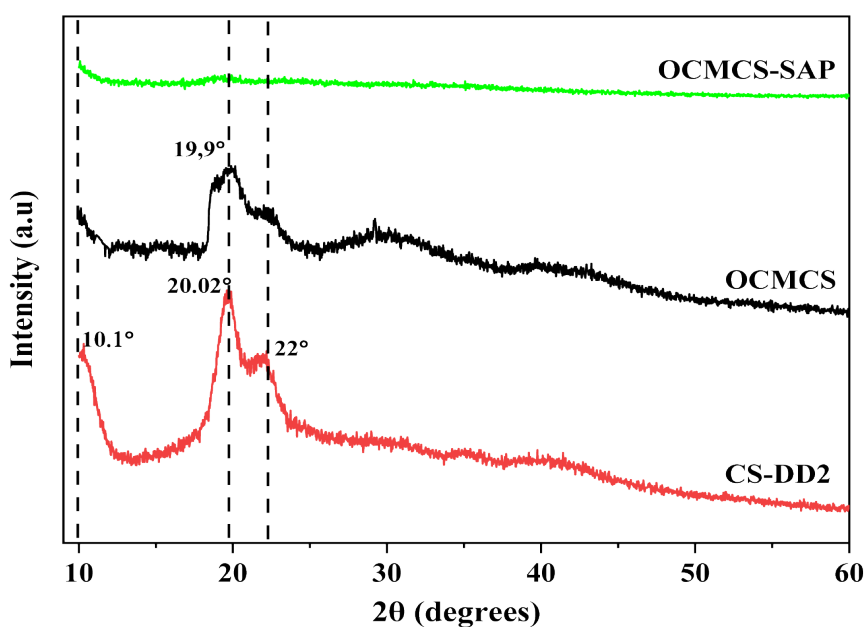


Figure 13. XRD spectra of CS-DD2, O-CMCS, and O-CMCS-SAP.

Thermogravimetric analysis (TGA) was used to analyze the material samples for thermal stability, and Figure 14 displays the mass loss of the material over various temperature ranges.

A two-stage decomposition mechanism illustrates the thermal breakdown of CS (Figure 14a). Around 100°C is the first stage; the mass loss of 13.12% in this temperature range is associated with the physical adsorption loss of water molecules. At 260-345°C, the second

stage of decrease takes place about 38.53%, and at 304°C, the rate of thermal breakdown peaks. The decomposition of the main chain of CS with rising temperature—that is, the oxidation, combustion, and decomposition of deoxidized CS units—could be the cause of this mass loss (46.81%).¹⁷

The thermal breakdown for the O-CMCS TGA diagram primarily occurred in three stages, at 170°C (9.17%), 293°C (33.58%),

and 530°C (57.25%) (Figure 14b). Because the initial thermal decomposition of O-CMCS was at 170°C, the second thermal decomposition interval peaked at 292°C lower than that of CS, and the third thermal decomposition interval reached 530°C, the temperature of the derivative was therefore worse than the decomposition temperature of CS when comparing the TGA diagrams of CS and O-CMCS. It was explained that the original crystal structure of CS was destroyed and the thermal stability of O-CMCS was decreased as a result of the high temperature and alkali breaking the macromolecular chains of CS during the carboxymethylation process.²³

Figure 14c displays the results of the thermogravimetric study of the O-CMCS-based superabsorbent polymer (O-CMCS-SAP). There were four spots of thermal deterioration found.

Due to the loss of water in the O-CMCS-SAP, the weight loss of 12.35% below 220°C (peaking at 195°C) was the initial step. The breakdown of the polymer's internal groups and branches following graft polymerization resulted in the second stage, which took place between 220°C and 300°C (peaking at 260°C), and the reduction of 20.24%. The third stage was brought on by the breakdown of the carboxymethyl group in the graft copolymer between 300°C and 440°C (peaking at 402°C, 40.64%). The weight loss at this high temperature range suggested that this was the temperature range for polymer chain degradation, as the final reduction stage took place above 440°C (peaking at 483°C, 26.77%).²³ The chitosan-derived grafted superabsorbent (O-CMCS) was shown to have a comparatively high degradation temperature based on the results of thermogravimetric analysis (TGA).

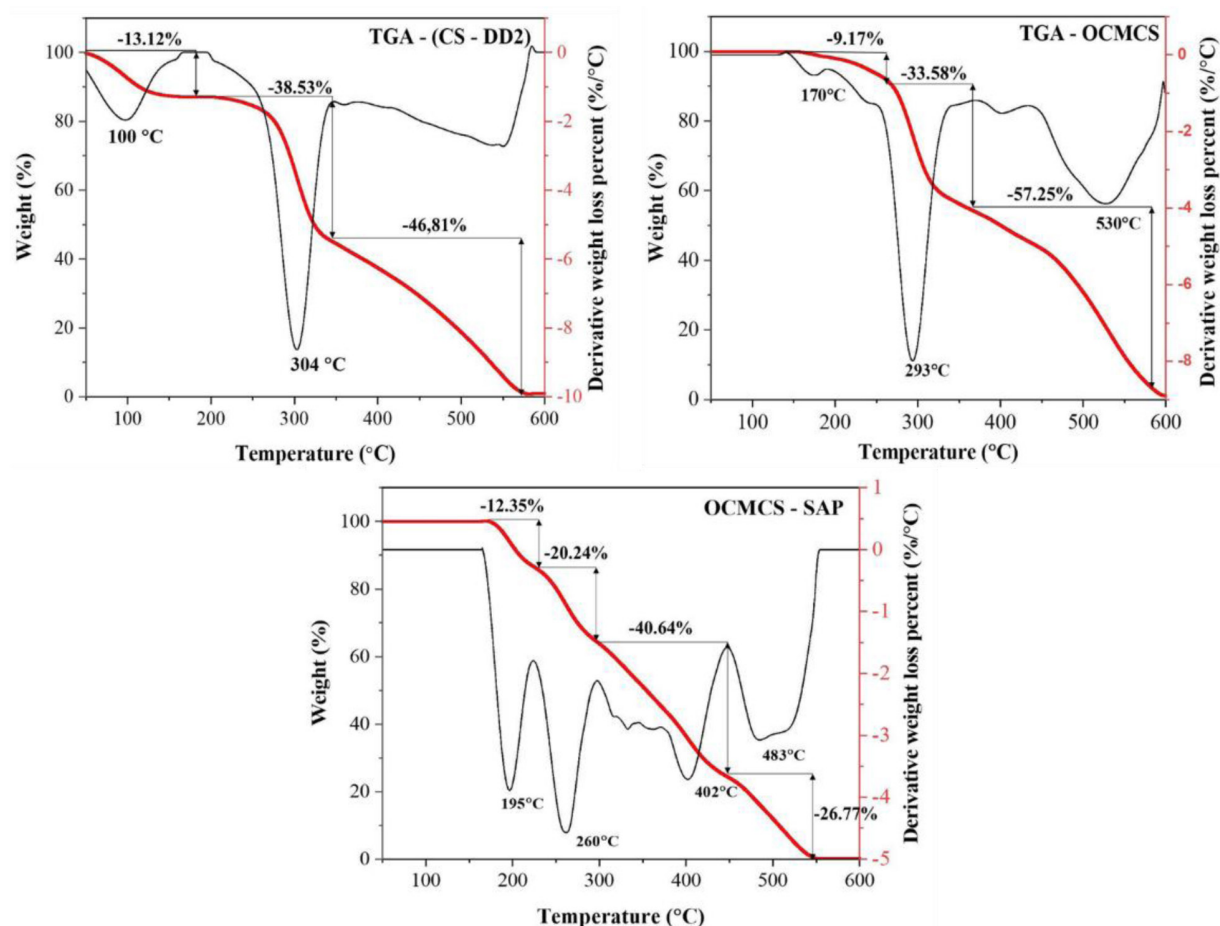


Figure 14. TGA results of CS-DD2, O-CMCS, and O-CMCS-SAP.

3.4. Examination of OCMCS-SAP's bio-degradability over time in a soil setting

In the soil environment, O-CMCS-SAP's bio-degradability over certain time intervals throughout four weeks is shown in Figure 15 and Table 1. As demonstrated throughout the course of the four weeks of the study, the mass of O-CMCS-SAP samples generally tends to increase slightly in the first week and after decrease significantly to the end of experiment for both unadsorbed and adsorbed states. The reason for this was that the modified chitosan has many functional groups and they easily associated with ions presented soil such as Ca^{2+} , Mg^{2+} , PO_4^{3-} . However, after the first week, the material was reduced more appreciably. This is because of the influence of the soil's micro-organisms and the temperature environment.

These elements have helped to create the conditions necessary for the substance to break down in the soil. The biodegradability of the material rose more than it had during the survey weeks. In particular, the dried sample began to decompose in the second week, and lost 35% compared to its initial mass at the end of the test, but the swollen sample started the biodegradation cycle in the second week and decomposed roughly 24% of its starting mass (5 g) by the fourth week. Even though the arid soil survey period was brief, it was sufficient for the material to continue biodegrading in an arid environment while still guaranteeing the ability to supply water to plants under challenging environmental circumstances. This is seen by the material's mass not declining too rapidly over time.

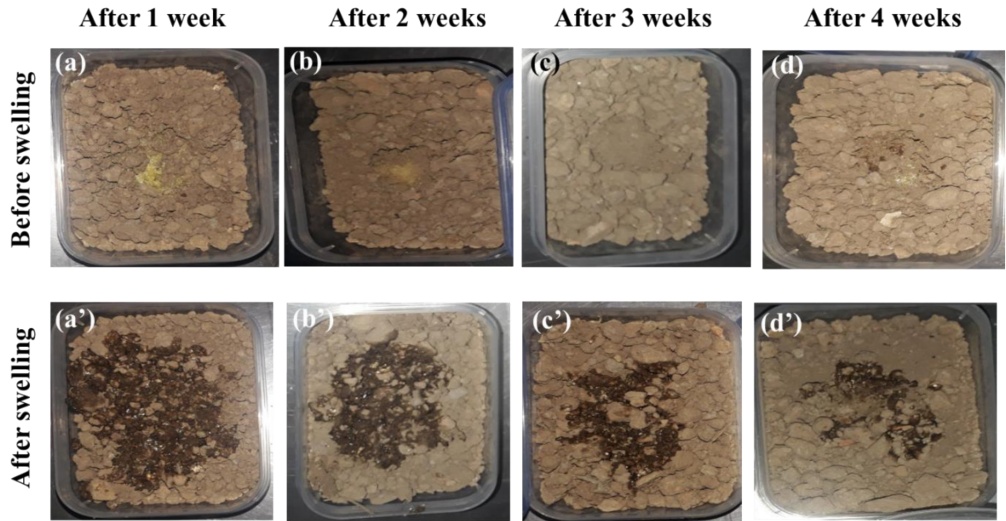


Figure 15. O-CMCS-SAP samples in dry soil for biodegradation before and after swelling at different time intervals.

Table 1. Experimental results of biodegradability of O-CMCS-SAP on dry soil before and after adsorption at different time intervals.

	After 1 week	After 2 weeks	After 3 weeks	After 4 weeks
for O-CMCS-SAP sample before adsorption				
Initial mass (g)	0.20	0.20	0.20	0.20
Final mass (g)	0.23	0.21	0.18	0.13
Biodegradability (%)	-	-	10	35
for O-CMCS-SAP sample after adsorption				
Initial mass (g)	5.0	5.0	5.0	5.0
Final mass (g)	5.3	4.9	4.5	3.8
Biodegradation (%)	-	7.55	10	24

The moist soil sample (humidity of 28%) was also surveyed for four weeks, just like the dry soil sample. Figure 16 displays the experimental soil samples, and Table 2 illustrates that its biodegradability. Similar to the infertile soil experiment, the material showed a mass increase during the first 2 weeks followed by a rapid mass loss until the forth week of the experiment. The dry material's tendency to increase in mass when in a humid environment with water present is caused by the material absorbing a certain amount of water into its structure. In contrast, the dry sample in the waterless soil environment as previously analyzed does not exhibit this phenomenon. The expanded sample also demonstrated the ability to bind to mineral components in moist soil owing to the active groups. It can be seen that O-CMCS-SAP material not only has the ability to absorb/release water but also the ability to load/deliver nutrients to provide for plants. The soil with pH between 6 - 7 was a suitable environment for microorganisms and enzymes

to grow that is appropriate for biodegradation of material increases with time. However, they are also susceptible to inactivation and reduced performance in alkaline environments when compared to SAP materials derived from synthetic polymers.²⁴ For moist or even dry soil, the material after adsorption is therefore fully biodegradable, but the initial particles prior to adsorption are still biodegradable but require more time.

The findings of the study have shown that O-CMCS-SAP material has better benefits than synthetic superabsorbent materials.^{2,25} Despite the short survey period, it is sufficient to determine the material's gradual mass decrease. The fact that they degrade but not too quickly and that they still retain some water in their structure shows that even though the material is breaking down over time, it still guarantees the plant's access to water during its growth and development phase.

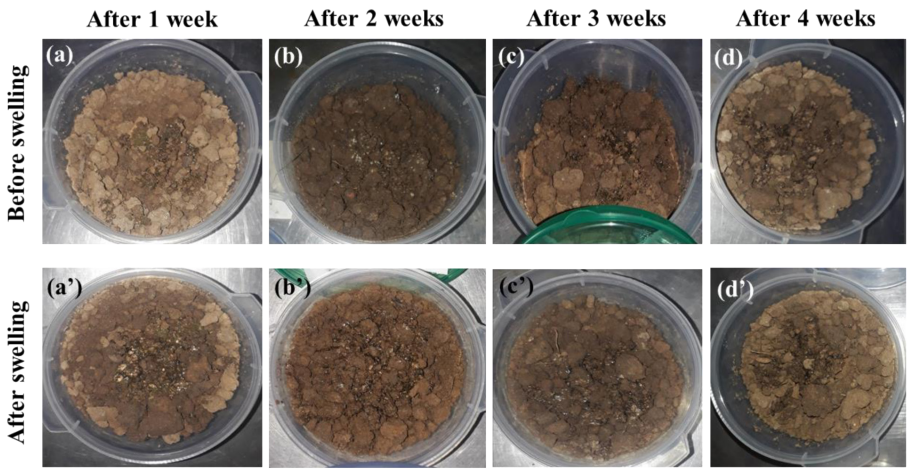


Figure 16. O-CMCS-SAP samples in moist soil (humidity 28%) for biodegradation before and after swelling at different time intervals.

Table 2. Experimental results of biodegradability of O-CMCS-SAP on moist soil (humidity 28%) before and after adsorption at different time intervals.

	After 1 week	After 2 weeks	After 3 weeks	After 4 weeks
for O-CMCS-SAP sample before adsorption				
Initial mass (g)	0.20	0.20	0.20	0.20
Final mass (g)	2.20	2.32	2.26	1.74
Biodegradability (%)	-	-	2.59	25
for O-CMCS-SAP sample after adsorption				
Initial mass (g)	5.00	5.00	5.00	5.00
Final mass (g)	5.50	5.20	4.84	3.87
Biodegradability (%)	-	-	3.2	22.6

3.5. Comparison of growth between two plants with and without O-CMCS-SAP materials

The growth of two identical chili plants was compared over time with and without O-CMCS-SAP. O-CMCS-SAP particles were dissolved in water and used to water the first plant (Figure 17a),

which was left unwatered for the duration of the study. During the first 10 days, the second chili plant (Figure 17b) received merely watering and was not treated with O-CMCS-SAP. The observed growth of the two plants is shown in Figure 17.

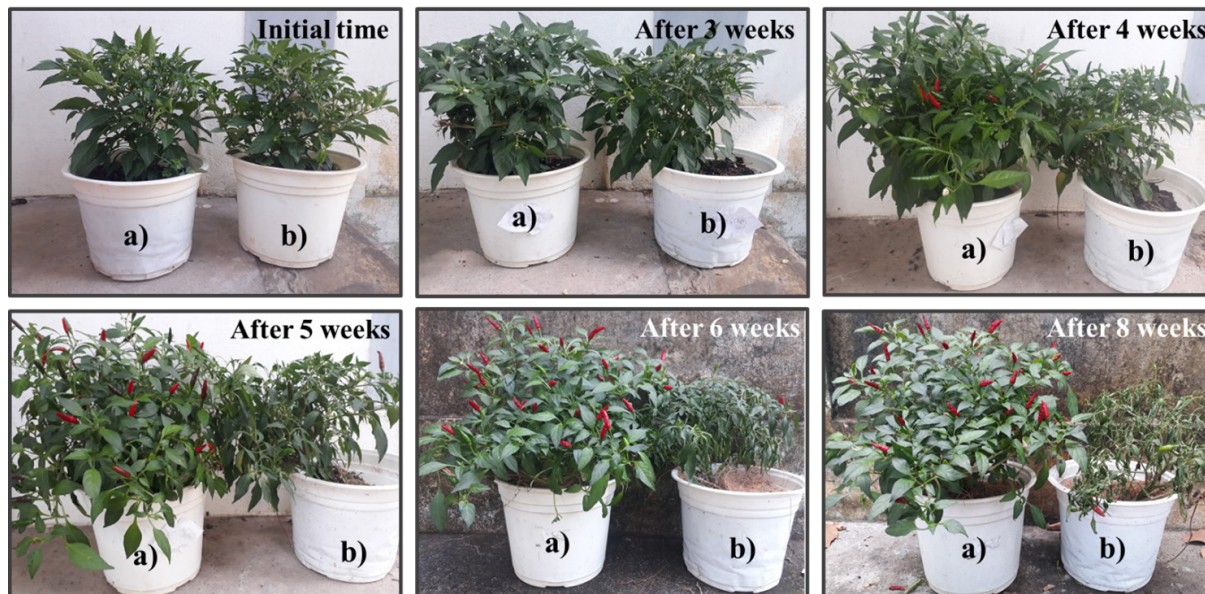


Figure 17. The distinction between two plants that (a) with and (b) without O-CMCS-SAP.

The initial weeks of observation showed no discernible change. In the two chili plants from the fourth week, the development was more noticeable. In comparison to the second chili plant (Figure 17b), the original chili plant (Figure 17a) grew taller, produced more fruits, and had more leaves. In contrast, the second plant (Figure 17b) did not grow taller and produced relatively few fruits. The results were more accurate by weeks five and six when the first chili plant produced a lot of fruit and continued to expand as the number of leaves increased. In contrast, the second chili plant produced nearly no fruit since it was not receiving enough water to grow and its leaves were wilting. The second plant was in danger of dying if it was not given water by the eighth week, even though the first plant was still growing and developing rapidly. The O-CMCS-SAP superabsorbent material's ability to adsorb and desorb water for over eight weeks was validated in this experiment, which helped to alleviate the issue of dry soil planting

areas and cut down on irrigation frequency. Additionally, study could be expanded to include the adsorption of other nutrients for plants.

4. CONCLUSIONS

The study has successfully utilized by-products in the seafood processing industry (shrimp head and body shells), which are prone to environmental pollution, to manufacture chitosan using a two-step deacetylation procedure with a high level of deacetylation. The first deacetylated chitosan had a deacetylation rate of 79.68%, whereas the second deacetylated chitosan had a high deacetylation rate of up to 91.48%. Moreover, the study successfully overcomes chitosan's poor solubility in neutral and alkaline pH environments by attaching functional groups to the main chain of chitosan, increasing chitosan's solubility in pH>6.5 environments, and then grafting the chitosan derivative with acrylic acid (AA) to create a superabsorbent polymer material from O-carboxymethyl chitosan

(O-CMCS-SAP). When O-CMCS-SAP material was synthesized from O-carboxymethyl and acrylic acid in the presence of the cross-linking agent glutaraldehyde, it attained a maximum adsorption capacity of 658.16 g/g after 15 days of immersion in water. Simultaneously, the optimal parameters for synthesizing materials with optimal adsorption capability were 2% potassium persulfate concentration, an O-CMCS:AA ratio of 1:8, and 2% glutaraldehyde concentration.

The capacity to biodegrade (as shown by four weeks of testing) and retain water to supply the plant during its growth is a key benefit of the superabsorbent material made from O-carboxymethyl chitosan. Additionally, the study was successful in analyzing the formation and growth of two identical chili plants. Because the plants grew taller and produced more fruit, the results demonstrated that the chili plants utilizing O-CMCS-SAP had much higher growth and development than those not using O-CMCS-SAP. After eight weeks of testing, the unwatered plants showed wilting leaves and extremely low development. Based on all of the experimental findings, the O-CMCS-SAP material has been successfully synthesized and has the potential to be applied to improve arid lands.

REFERENCES

1. Y. Chen, H. Tan. Crosslinked carboxymethylchitosan-g-poly (acrylic acid) copolymer as a novel superabsorbent polymer, *Carbohydrate Research*, **2006**, 341(7), 887-896.
2. S. Fang, G. Wang, R. Xing, X. Chen, S. Liu, Y. Qin, K. Li, X. Wang, R. Li, P. Li. Synthesis of superabsorbent polymers based on chitosan derivative graft acrylic acid-co-acrylamide and its property testing, *International Journal of Biological Macromolecules*, **2019**, 132, 575-584.
3. V. K. Nguyen, T. T. Nguyen, T. D. Nguyen, T. T. H. Pham, T. T. Pham, V. T. Tran, T. H. Nguyen, T. L. P. Nguyen. Superabsorbent polymers - an innovative solution for improving water usage efficiency and agriculture productivity, *Vietnam Journal of Science and Technology*, **2022**, 60(3), 314-332.
4. N. Nhan. God of water for plants, *Science and Life Newspaper*, **2006**, 103.
5. S. C. Sutradhar, M. M. R. Khan, M. M. Rahman, N. C. Dafadar. The synthesis of superabsorbent polymers from a carboxymethylcellulose/ acrylic acid blend using gamma radiation and its application in agriculture, *Journal of Physical Science*, **2015**, 26(2), 23-39.
6. A. Narayanan, R. Kartik, E. Sangeetha, R. Dhamodharan. Super water absorbing polymeric gel from chitosan, citric acid and urea: synthesis and mechanism of water absorption, *Carbohydrate Polymers*, **2018**, 191, 152-160.
7. N. Neamjan, R. Wiwattanankul, K. Hiangrat, K. Matkaran, Y. Na-iam, M. Sakulsombat, K. Sriroth. Preparation of superabsorbent polymer from sugarcane bagasse via extrusion process, *Sugar Tech*, **2019**, 21(2), 296-300.
8. B. A. Yassine, M. Bezbiz, L. Belachemi, C. Moreau, C. Garnier, C. Jonchere, H. Benyoucef, B. Cathala, H. Kaddami. Preparation of superabsorbent composite (s) based on dialdehyde cellulose extracted from banana fiber waste, *Carbohydrate Polymers*, **2024**, 343, 122504.
9. J. Zhang, S. Bai, J. Lyu, X. Guan. Synthesis of a spherical starch-based superabsorbent polymer and its influence on the microstructure of hardened cement paste, *Composites Part B: Engineering*, **2025**, 297, 112310.
10. X. Guan, J. Zhang, S. Zhao. Design, synthesis and characterization of a starch-based superabsorbent polymer and its impact on autogenous shrinkage of cement paste, *Construction and Building Materials*, **2024**, 415, 134986.
11. S. Sahoo. Chitin/chitosan based superabsorbent polymers. *Bio-based superabsorbents: recent trends, types, applications and recycling*, Springer Nature Singapore, Singapore, 2023.

12. M. Jafari, G. R. Najafi, M. A. Sharif, Z. Elyasi. Superabsorbent polymer composites derived from polyacrylic acid: design and synthesis, characterization, and swelling capacities, *Polymers and Polymer Composites*, **2021**, 29(6), 733-739.
13. E. Czarnecka, J. Nowaczyk. Synthesis and characterization superabsorbent polymers made of starch, acrylic acid, acrylamide, poly (Vinyl alcohol), 2-hydroxyethyl methacrylate, 2-acrylamido-2-methylpropane sulfonic acid, *International Journal of Molecular Sciences*, **2021**, 22(9), 4325.
14. M. Kurniasih, P. Purwati, D. Hermawan, M. Zaki. Optimum conditions for the synthesis of high solubility carboxymethyl chitosan, *Malaysian Journal of Fundamental and Applied Sciences*, **2014**, 10(4), 189-194.
15. P. V. Du, N. T. K. Thuong, N. V. Thom. The extraction and antibacterial ability of water-soluble chitosan from white leg shrimp shell (*Litopenaeus vannamei*), *Can Tho University Journal of Science*, **2020**, 56(2), 136-145.
16. R. R. Mohamed, N. A. Rizk, B. M. A. E. Hady, H. M. Abdallah, M. W. Sabaa. Synthesis, characterization and application of biodegradable crosslinked carboxymethyl chitosan/poly (ethylene glycol) clay nanocomposites, *Journal of Polymers and the Environment*, **2017**, 25, 667-682.
17. S. Fang, G. Wang, P. Li, R. Xing, S. Liu, Y. Qin, H. Yu, X. Chen, K. Li. Synthesis of chitosan derivative graft acrylic acid superabsorbent polymers and its application as water retaining agent, *International Journal of Biological Macromolecules*, **2018**, 115, 754-761.
18. H. F. Harrar, N. Aiouaz, N. Dairi, A. S. H. Hamou. Preparation of chitosan-g-poly (acrylamide)/montmorillonite superabsorbent polymer composites: studies on swelling, thermal, and antibacterial properties, *Journal of Applied Polymer Science*, **2014**, 131(1), 39747.
19. Y. Chen, Y. F. Liu, H. M. Tan, J. X. Jiang. Synthesis and characterization of a novel superabsorbent polymer of N, O-carboxymethyl chitosan graft copolymerized with vinyl monomers, *Carbohydrate Polymers*, **2009**, 75(2), 287-292.
20. H. Ge, S. Wang. Thermal preparation of chitosan-acrylic acid superabsorbent: optimization, characteristic and water absorbency, *Carbohydrate polymers*, **2014**, 113, 296-303.
21. N. M. B. Ladeira, C. L. Donnici, J. P. Mesquita, F. V. Pereira. Preparation and characterization of hydrogels obtained from chitosan and carboxymethyl chitosan, *Journal of Polymer Research*, **2021**, 28(9), 335.
22. Y. Chen, Y. Yang, Q. Liao, W. Yang, W. Ma, J. Zhao, X. Zheng, Y. Yang, R. Chen. Preparation, property of the complex of carboxymethyl chitosan grafted copolymer with iodine and application of it in cervical antibacterial biomembrane, *Materials Science and Engineering: C*, **2016**, 67, 247-258.
23. M. Behrouzi, P. N. Moghadam. Synthesis of a new superabsorbent copolymer based on acrylic acid grafted onto carboxymethyl tragacanth, *Carbohydrate Polymers*, **2018**, 202, 227-235.
24. C. Zhang, J. V. G. Meza, K. Zhou, J. Liu, S. Song, M. Zhang, D. Meng, J. Chen, L. Xia, H. Xiheng. Superabsorbent polymer used for saline-alkali soil water retention, *Journal of the Taiwan Institute of Chemical Engineers*, **2023**, 145, 104830.
25. E. M. Ahmed, M. A. Zahran, F. S. Aggor, S. Elhady, S. S. Nada. Synthesis and swelling characterization of carboxymethyl cellulose-g-poly (acrylic acid-co-acrylamide) hydrogel and their application in agricultural field, *International Journal of ChemTech Research*, **2016**, 9(8), 270-281.



© 2025 by the authors. This Open Access Article is licensed under the Creative Commons Attribution-NonCommercial 4.0 International (CC BY-NC 4.0) license (<https://creativecommons.org/licenses/by-nc/4.0/>).

Các bóng bán dẫn GAAFET và vật liệu 2D cho công nghệ dưới 3 nm: hiệu năng, thách thức chế tạo và chiến lược tích hợp

Huỳnh Công Tú^{1,*}, Hoàng Anh Phương²

¹Phòng Đào tạo, Trường Đại học Quy Nhơn, Việt Nam

²Khoa Kỹ thuật cơ sở, Trường Sĩ quan Không quân, Việt Nam

Ngày nhận bài: 10/04/2025; Ngày sửa bài: 07/06/2025;

Ngày nhận đăng: 11/07/2025; Ngày xuất bản: 28/10/2025

TÓM TẮT

Bài báo này nghiên cứu quá trình chuyển đổi từ FinFETs sang Transistor Bao Quanh Cổng (Gate-All-Around Field-Effect Transistors - GAAFETs) như một bước tiến chiến lược cho các nút công nghệ CMOS vượt ngưỡng 3 nm, với trọng tâm là các cải tiến hiệu suất, thách thức chế tạo và chiến lược tích hợp. Chúng tôi thực hiện phân tích so sánh các kiến trúc transistor tiên tiến - bao gồm FinFET 3 nm của TSMC, GAAFET dạng tấm nano 3 nm của Samsung (MBCFETTM), và RibbonFET 20A sắp ra mắt của Intel – nhằm làm nổi bật khả năng điều khiển điện trường và dòng dẫn vượt trội của GAAFETs. Vai trò của các vật liệu tiên tiến như SiGe, các hợp chất III-V, và chất bán dẫn hai chiều (ví dụ: MoS- đơn lớp và WSe-) được phân tích trong bối cảnh mở rộng khả năng thu nhỏ xuống dưới 1 nm, đặc biệt là với khả năng giảm thiểu hiệu ứng kênh ngắn. Chúng tôi cũng xem xét các thách thức chế tạo chính liên quan đến công nghệ dưới 3 nm, bao gồm giới hạn của quang khắc EUV, nhu cầu kiểm soát quy trình ở cấp nguyên tử và chi phí sản xuất leo thang. Bên cạnh đó, các kỹ thuật tích hợp dị thể như thiết kế chiplet và xếp chồng 3D được đề xuất như những cách tiếp cận bổ sung để duy trì xu hướng cải thiện hiệu suất. Cuối cùng, chúng tôi thảo luận về các kiến trúc thiết bị sau GAAFET đầy triển vọng - bao gồm CFETs (Transistor bổ sung) và VTFETs (Transistor truyền dọc) - nhấn mạnh nhu cầu đổi mới liên ngành để duy trì quỹ đạo của định luật Moore. Những kết quả thu được cung cấp một góc nhìn toàn diện về việc thu nhỏ thiết bị bán dẫn trong thời đại “<3 nm”, với sự cân bằng giữa hiệu suất, hiệu quả năng lượng và khả năng sản xuất.

Từ khóa: GAAFET, FinFET, vật liệu hai chiều, quang khắc EUV, định luật Moore.

*Tác giả liên hệ chính.

Email: huynhcongту@qnu.edu.vn

GAAFETs and 2D materials for sub-3 nm nodes: performance, fabrication challenges, and integration strategies

Huynh Cong Tu^{1,*}, Hoang Anh Phuong²

¹Training office, Quy Nhon University, Vietnam

²Faculty of Basic Engineering, Air Force Officer Academy, Vietnam

Received: 10/04/2025; Revised: 07/06/2025;

Accepted: 11/07/2025; Published: 28/10/2025

ABSTRACT

This paper investigates the transition from FinFETs to Gate-All-Around Field-Effect Transistors (GAAFETs) as a strategic advancement for CMOS technology nodes beyond 3 nm, with a focus on performance enhancements, fabrication challenges, and integration strategies. We conduct a comparative analysis of state-of-the-art transistor architectures—namely TSMC’s 3 nm FinFET, Samsung’s 3 nm nanosheet GAAFET (MBCFET™), and Intel’s upcoming 20A RibbonFET—to highlight the superior electrostatic control and drive current offered by GAAFETs. The role of emerging materials such as SiGe, III–V compounds, and two-dimensional (2D) semiconductors (e.g., monolayer MoS₂ and WSe₂) is examined in the context of extending transistor scaling into the sub-1 nm regime, particularly with respect to their potential to mitigate short-channel effects. We further address key fabrication challenges associated with sub-3 nm technologies, including the limitations of extreme ultraviolet (EUV) lithography, the need for atomic-scale process control, and escalating production costs. In addition, we explore heterogeneous integration techniques, such as chiplet-based design and 3D stacking, as complementary approaches to sustain performance scaling. Finally, we discuss prospective post-GAAFET device architectures—including Complementary FETs (CFETs) and Vertical Transport FETs (VTFETs)—emphasizing the necessity of cross-disciplinary innovation to uphold the trajectory of Moore’s Law. The findings present a comprehensive perspective on semiconductor scaling in the “<3 nm” era, balancing trade-offs between performance, energy efficiency, and manufacturability.

Keywords: GAAFET, FinFET, 2D materials, EUV lithography, Moore’s law.

1. INTRODUCTION

For several decades, CMOS technology scaling followed a consistent trajectory of planar transistor miniaturization. However, by the 22–20 nm technology node around 2011, planar MOSFETs began to encounter severe short-channel effects and increasing leakage currents, which ultimately limited further downscaling.^{1,2} The introduction of the tri-dimensional Fin

Field-Effect Transistor (FinFET) marked a pivotal transformation in transistor architecture, restoring electrostatic control and enabling continued scaling. Intel commercialized the first FinFET at the 22 nm node in 2012, followed by major foundries such as TSMC and Samsung at the 16/14 nm nodes.^{3,4} In a FinFET, the channel adopts a vertical fin structure, and the gate wraps around three sides of the fin, unlike planar FETs

*Corresponding author.

Email: huynhcongту@qnu.edu.vn

where the gate contacts only one side. This geometry significantly enhances gate control over the channel, reducing off-state leakage and facilitating transistor scaling down to the 10 nm, 7 nm, and 5 nm nodes while maintaining sufficient drive current and energy efficiency.⁵

As FinFETs approach gate lengths of approximately 5 nm, their inherent physical limitations have become more pronounced. The requirement for a minimum fin width and spacing imposes a quantized and non-continuous channel width, thereby restricting fine-grained width control and drive-current tuning at ultra-scaled dimensions. Moreover, ultra-narrow fins are more susceptible to performance degradation due to increased series resistance and process variability. While multiple fins can be used in parallel to boost drive current, practical constraints on fin count and layout density limit the extent to which this can be scaled. These factors collectively lead to diminishing returns in performance and rising leakage as FinFETs are extended beyond the ~5 nm threshold.^{5,6} Consequently, the FinFET architecture is approaching a fundamental scaling barrier, prompting the development of new transistor structures capable of sustaining Moore's Law into the sub-3 nm regime.^{7,8}

Gate-All-Around Field-Effect Transistors (GAAFETs) – realized in practice through nanosheet or nanowire architectures – have emerged as the leading candidates for next-generation nodes at 3 nm, 2 nm, and beyond.^{6,9} In a GAAFET, the gate completely surrounds the semiconductor channel on all four sides, offering superior electrostatic control compared to FinFETs, which only provide tri-gate coverage. This fully-surrounding gate configuration eliminates exposed channel regions that contribute to leakage in FinFETs. Figure 1 illustrates this structural progression: from planar FETs (gate on one side), to FinFETs (three-sided gate), to GAAFETs (fully wrapped gate). By eliminating the open top of the channel found in FinFETs, GAAFETs more effectively

suppress short-channel effects, resulting in sharper transistor switching and significantly reduced off-state leakage.

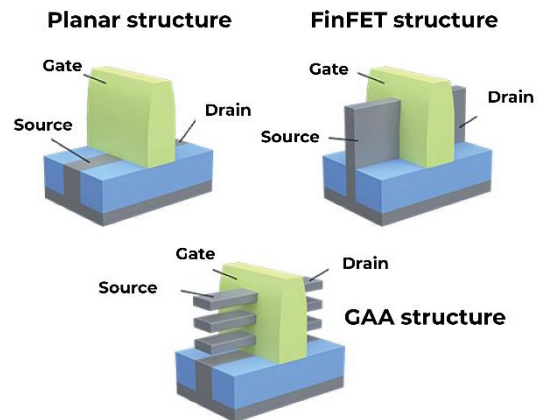


Figure 1. Schematic comparison of transistor architectures: (top-left) planar MOSFET with single-side gate control; (top-right) FinFET with tri-gate coverage; (bottom) gate-all-around (GAA) nanosheet FET in which the metal gate fully surrounds stacked Si/SiGe channels, enabling superior electrostatic control at sub-3 nm nodes. All renderings are to scale for a 16 nm gate length and ~45 nm fin/nanosheet height.

Another key advantage of nanosheet-based GAAFETs is their ability to continuously and independently adjust channel width. In FinFETs, the effective channel width is quantized, as it can only be increased in discrete steps by adding additional fins. In contrast, GAAFETs allow for the stacking of multiple horizontal nanosheet channels, each with customizable width. This architecture enables circuit designers to finely tune drive strength by selecting the number and dimensions of nanosheets.^{10,11} Such flexibility allows for optimal balancing of performance and power consumption at the individual device level. Unlike FinFETs—where an intermediate configuration (e.g., 2.5 fins) is physically unattainable – GAAFETs eliminate this granularity constraint, simplifying standard cell library design and potentially improving circuit density and operating speed.

Initial findings from both academia and industry indicate that GAAFETs outperform FinFETs in advanced technology nodes. For

example, CEA-Leti demonstrated a seven-layer stacked nanosheet GAAFET with nearly 3× higher drive current compared to a conventional two-layer counterpart, emphasizing the benefits of wider effective channel widths through vertical stacking.¹² Likewise, Samsung Electronics reported that its first-generation 3 nm GAA process (MBCFET™) achieved up to 45% reduction in power consumption and a 23% increase in performance compared to its 5 nm FinFET process.¹³ These substantial improvements, however, come at the cost of increased fabrication complexity. Stacking nanosheets requires extremely precise atomic-level process control, advanced etching techniques, and in some cases the integration of alternative channel materials such as SiGe to apply strain or engineer specific shapes. As a result, while GAAFETs offer clear scaling advantages, they also present initial challenges in terms of cost and manufacturability. Nevertheless, the transition to GAAFETs is widely regarded as essential for maintaining scaling momentum.

In parallel with structural innovations, researchers are exploring new channel materials to further extend transistor scaling. Silicon, long the cornerstone of the semiconductor industry, faces intrinsic physical limitations at atomic scales, including mobility degradation and quantum confinement effects. Two-dimensional (2D) semiconductors – such as monolayer transition metal dichalcogenides (TMDs) like MoS₂ and WSe₂ – have emerged as promising alternatives due to their atomically thin nature, excellent electrostatics, and favorable carrier mobilities. These materials lack dangling bonds on their surfaces, enabling near-ideal interfaces with gate dielectrics even at sub-nanometer thicknesses.¹⁴ As a result, they offer a viable pathway for scaling gate lengths below 5 nm without incurring severe short-channel effects as observed in silicon-based transistors. Experimental 2D-FETs have demonstrated high on/off ratios and low leakage currents at

channel lengths as short as ~1 nm, indicating their potential to extend Moore's Law beyond the material limitations of silicon.¹⁴ However, achieving large-scale, reliable integration of 2D materials into mainstream fabrication processes remains a significant challenge, as further discussed in Section 4.

In addition to device-level advancements, system-level innovations are being pursued to complement and extend the benefits of transistor scaling. As conventional scaling becomes increasingly difficult and cost-prohibitive, heterogeneous integration techniques – such as chiplet-based architectures and 3D stacking – are gaining traction.¹⁵ These strategies aim to improve overall system performance through advanced packaging, for example by dividing a system-on-chip into multiple function-specific chiplets that can be fabricated at optimal nodes and then interconnected, or by vertically integrating logic and memory dies to minimize interconnect delay and energy loss. Even as transistor scaling slows, such integration techniques offer a viable path toward continued performance and efficiency gains.

This paper provides a comprehensive analysis of GAAFET technology and its role in sub-3 nm semiconductor scaling. The key contributions of this work include:

- Comparative evaluation of FinFETs and GAAFETs: We assess performance parameters (e.g., drive current, transconductance), leakage control (e.g., subthreshold swing, DIBL), and design flexibility to highlight the advantages of GAAFETs at advanced nodes.
- Industry case studies: We examine leading-edge 3 nm processes from TSMC (FinFET) and Samsung (GAAFET), and evaluate Intel's roadmap to 2 nm with RibbonFET, focusing on reported gains in power and performance.
- Exploration of 2D materials: We investigate the potential of 2D semiconductors to support scaling into the sub-1 nm regime,

discussing their benefits and the technical barriers to mass production.

- Fabrication and integration challenges: We analyze key manufacturing constraints at advanced nodes, including EUV lithography limitations, atomic-level variability, and rising wafer costs, and explore heterogeneous integration as a complementary strategy.

- Prospects for future architectures: We consider emerging options beyond GAAFETs—such as Complementary FETs (CFETs) and Vertical Transport FETs (VTFETs)—and emphasize the need for cross-disciplinary research spanning materials, processing, and circuit design to realize next-generation devices.

By addressing these dimensions, this study outlines a strategic roadmap for innovation in semiconductor technologies beyond the 3 nm node, emphasizing how performance, energy efficiency, and manufacturability trade-offs can be effectively navigated over the coming decade.

2. COMPARATIVE ANALYSIS OF FINFETS AND GAAFETS

The transition from FinFETs to GAAFETs is fundamentally motivated by the latter's superior electrostatic control and enhanced design flexibility. This section presents a detailed comparison between FinFET and GAAFET devices across key performance metrics to quantitatively assess these advancements.

2.1. Performance enhancements and electrostatic control

Drive current (ion) and width scaling:

FinFETs facilitated sub-10 nm technology scaling primarily due to their improved gate control compared to planar FETs. However, their drive current capability becomes limited near the 5 nm node due to the quantized nature of channel width imposed by fin geometry.^{5,6} A FinFET's effective width is approximated as twice the fin height plus the top width, multiplied by the number of fins. This discrete quantization

restricts designers to increase drive current only in fixed increments by adding fins. Figure 2 provides a 3D depiction and cross-sectional comparison of the GAAFET structure.

In contrast, GAAFETs enable continuous width scaling. Through the stacking of multiple nanosheets, the effective channel width – and consequently I_{on} – can be finely tuned to meet performance demands.¹¹ Empirical studies have demonstrated that multi-stack GAAFETs can deliver up to a $3\times$ increase in drive current compared to equivalent single- or dual-fin FinFETs, owing to their broader effective channels.¹² This directly translates to enhanced switching speeds and greater current-driving capability in logic cells.

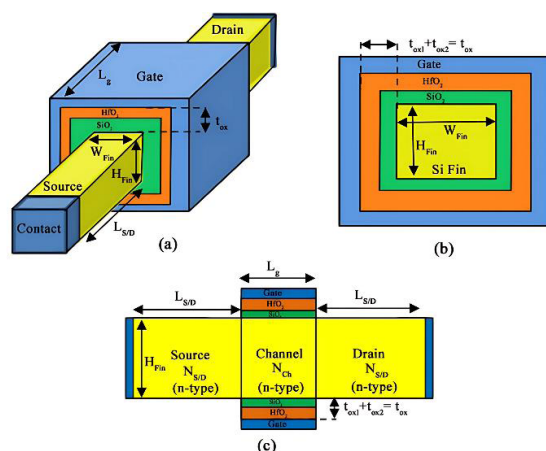


Figure 2. GAAFET nanosheet FET schematics: (a) 3D perspective showing n-type source/channel/drain regions, Si_{fin} width W_n and height H_n , HfO₂ gate-oxide of thickness t_{ox} surrounding the Si_{fin}, metal gate, and source/drain extensions $L_{s/d}$. (b) Top-view cross-section illustrating concentric layers – Si_{fin} (yellow), SiO₂ liner (green), HfO₂ gate-oxide (orange, $t_{ox1} + t_{ox2} = t_{ox}$), and metal gate (blue). (c) Side cross-section perpendicular to the fin, showing channel region (L_g), source/drain doping ($N_{s/d}$), and gate stack. All dimensions (L_g , $L_{s/d}$, W_n , H_n , t_{ox}) require atomic-scale (± 0.2 nm) control at sub-3 nm nodes.

Transconductance (g_m) and gate control:

Transconductance ($g_m = \partial I_D / \partial V_G$) quantifies the gate's ability to modulate channel charge. Thanks to their gate-all-around structure,

GAAFETs achieve stronger gate-channel coupling capacitance and reduced charge sharing, resulting in significantly higher g_m than FinFETs at equivalent dimensions.¹⁶ With the gate enveloping the channel from all sides, even small variations in gate voltage induce greater charge modulation. Higher g_m not only improves digital switching speed but also benefits analog and RF applications by delivering higher gain and bandwidth. Specifically, recent 3 nm GAAFET simulations using the GT3 open-source PDK indicate marked improvements in drive current and subthreshold swing compared to junctionless transistor designs, further confirming GAAFET’s advantages at extreme scaling.^{17,18} Complementing these findings, TCAD work at the projected 2 nm node shows that both lateral- and vertical-nanosheet GAAFETs maintain superior I_{on}/I_{off} and short-channel control over equivalent nanowire and FinFET geometries, even when effective widths are swept from 18 nm to 54 nm.¹⁹

Subthreshold swing (SS) and off-state leakage (I_{off}):

Subthreshold swing (SS) reflects the steepness of the transition from off to on state and is defined as the gate voltage required to change the drain current by one order of magnitude. The ideal thermal limit is ~60 mV/dec at room temperature. FinFETs at advanced nodes typically exhibit SS values in the 70 - 80 mV/dec range due to partial leakage from the fin top. In contrast, GAAFETs approach the theoretical limit more closely due to complete channel encapsulation.¹⁶ Practically, GAAFETs can reduce I_{off} by 30 - 50% compared to FinFETs at similar gate lengths and voltages.²⁰ This significant leakage reduction supports operation at lower threshold and supply voltages, contributing to lower static power consumption and improved energy efficiency.²¹

In Table 1, we summarize the adoption timelines and reported performance/power improvements of FinFET and GAAFET

technologies across leading manufacturers, illustrating how GAAFETs deliver superior electrostatics and power scaling at the 5 nm, 3 nm, and upcoming 2 nm nodes.

Table 1. Comparison of FinFET and GAAFET adoption across leading semiconductor manufacturers.

Company	Technology	Transistor Architecture	Performance & Power Gains	Production Timeline
TSMC	N3 (3 nm)	FinFET	+10 - 15% performance, -25-30% power (vs. 5 nm)	2022 - 2023
Samsung	3 nm	GAAFET (MBCFET™)	-45% power, +23% performance, -16% area (vs. 5 nm)	Early production in 2022, high volume in 2024
Intel	20A (~2 nm)	GAAFET (RibbonFET)	Expected benefits of nanosheet stacking	2024 (production), 2025 (commercial launch)
IBM	2 nm (prototype)	GAAFET (Nanosheet FET)	+45% performance, -75% power (vs. 7 nm)	2021 (research phase)

Drain-induced barrier lowering (DIBL) and short-channel effects:

DIBL measures the reduction in threshold voltage as the drain voltage increases – a critical indicator of short-channel control. GAAFETs, with their fully surrounding gate, exhibit superior electrostatic confinement, thereby minimizing the influence of drain electric fields. Consequently, GAAFETs achieve substantially lower DIBL than FinFETs at the same channel length.^{16,22} Lower DIBL ensures robust device operation at reduced supply voltages and mitigates premature conduction due to drain bias, which is crucial for ultra-low-power designs. In contrast, FinFETs exhibit worsening SS and DIBL performance as channel lengths shrink near 5 nm.⁶

Table 1 summarizes key performance parameters, demonstrating that GAAFETs outperform FinFETs in terms of I_{on} , g_m , SS, and DIBL at equivalent technology nodes. These improvements stem directly from the gate-all-around architecture, establishing GAAFETs as the preferred option for 5 nm, 3 nm, and beyond.

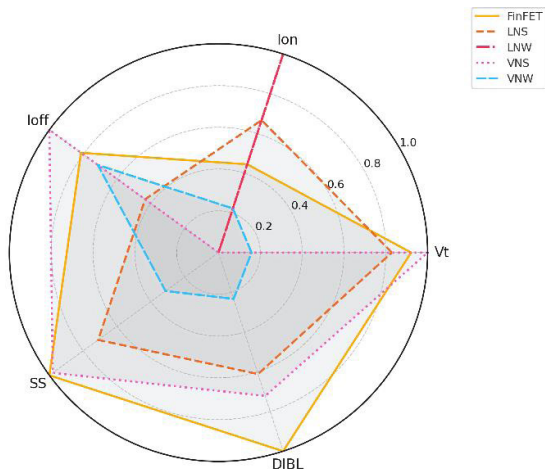


Figure 3. Radar chart comparing normalized performance metrics of FinFET and various GAAFET configurations at the 3 nm node: long nanosheet (LNS), long nanowire (LNW), vertical nanosheet (VNS), and vertical nanowire (VNW). Metrics include threshold voltage (V_t), on-current (I_{on}), off-current (I_{off}), subthreshold swing (SS), and DIBL, each scaled between 0 (worst) and 1 (best) for fair comparison.¹⁹

2.2. Leakage reduction and power efficiency

As technology nodes scale down, leakage current becomes a dominant factor in overall power consumption. FinFETs reduced leakage relative to planar FETs by introducing tri-gate control, but the top of the fin remains partially exposed. At nanoscale dimensions, this exposes the channel to fringe fields that can induce parasitic conduction - even when the device is off.²³ Figure 3 highlights the comparative plot of I_{on} , SS, and DIBL for FinFET vs. GAAFET.

GAAFETs address this by fully enclosing the channel. With gate coverage on all sides, electrostatic control becomes more uniform, eliminating weak leakage paths. Experimental results at 3 - 5 nm nodes show that GAAFETs can reduce subthreshold leakage (I_{off}) by approximately 30 - 50% compared to FinFETs under equivalent bias and geometry conditions.^{16,20} This translates to significantly lower standby power in digital systems and improved energy efficiency, especially in battery-powered and ultra-low-power applications.

Moreover, better subthreshold slope and reduced DIBL in GAAFETs enable lower threshold voltages and supply voltages without sacrificing switching behavior. Since dynamic power scales with CV^2f , the ability to lower V_{dd} while maintaining performance directly improves energy efficiency. For instance, Samsung's migration to GAAFETs at 3 nm allowed operation at reduced V_{dd} , yielding up to 45% total power reduction.¹³

In summary, GAAFETs offer substantial improvements in leakage control, enabling lower-voltage operation and reduced active and standby power. The ability to continuously scale nanosheet width further enhances efficiency and tuning capability compared to the quantized nature of FinFET fins. These attributes make GAAFETs especially well-suited for next-generation low-power and high-density logic circuits.²¹

2.3. Design flexibility and integration considerations

A frequently underappreciated advantage of GAAFETs lies in their design scalability. In FinFET-based standard cell libraries, drive strength is quantized by the number of fins per transistor. For example, scaling from a 2-fin to a 3-fin inverter results in a 50% width increase

and a proportional jump in capacitance. This granularity can complicate timing and power optimization,¹⁰ and necessitates creating and validating multiple variants (so-called FinFET binning), increasing design and verification overhead.

In contrast, GAAFETs provide continuous sizing flexibility. Designers can adjust the width of each nanosheet or the number of sheets lithographically to achieve the desired drive strength.¹¹ This allows library cells to be defined in finer width increments, reducing the number of distinct cells and enabling more precise timing/power trade-offs. For example, a designer could target a transistor width equivalent to 2.4 fins rather than being constrained to discrete integers. This granularity supports improved power distribution and timing closure in VLSI implementation.

From an integration standpoint, however, GAAFETs introduce new challenges. Their 3D stacked architecture requires careful routing and contact placement. Design rules may impose constraints on device placement to maintain nanosheet alignment or gate pattern uniformity. Furthermore, process modifications such as the use of SiGe for nanosheet release, or novel spacer and dielectric materials, must be co-optimized with physical design rules to ensure manufacturability and yield.

GAAFETs deliver superior device performance, better power efficiency, and enhanced design scalability compared to FinFETs. While their fabrication demands more complex integration efforts, their benefits are substantial enough to justify the industry-wide shift to GAAFETs at the 3 nm node and beyond. Table 3 provides a qualitative overview of technology readiness: FinFETs remain viable at 7 nm and 5 nm, but beyond this, GAAFETs offer a clear advantage in sustaining Moore’s Law through improved electrostatics, power scaling, and layout flexibility.

Table 2. Estimated cost and yield comparison between FinFET and GAAFET platforms at the 3 nm node.

Metric	FinFET (N3)	GAAFET (SF3E/20A)
Mask Count	78	88
High-NA EUV usage	No	Partial
Yield (first year)	70%	55%
Cost per cm ²	\$18.2	\$23.7
Cost per logic gate	0.85 p\$	1.12 p\$

2.4. Cost-benefit analysis for commercial implementation

While GAAFETs promise better electrostatic control and scaling, their manufacturing complexity leads to trade-offs in cost and yield. Das et al.⁸ report that MBCFET/GAAFET integration typically requires more photomasks than FinFET - roughly 88 vs. 78 masks at a given node - implying higher capital expenses. Scognamiglio et al.¹⁸ additionally confirm that process steps for advanced fin/stack alignment further increase fabrication time. Although detailed cost figures vary by foundry, the consensus is that initial GAAFET yield (~55 %) lags behind FinFET (~70 %) as reported by Ma et al.²⁴ Nevertheless, long-term gains in power efficiency and logic density - particularly when leveraging stress-engineered Si/SiGe channels²⁴ - may offset these upfront costs within one or two technology generations. Table 2 compares key metrics between FinFET and GAAFET technologies, including mask count, EUV exposure steps, and yield rates.

3. 3 NM AND 2 NM NODE DEPLOYMENTS BY INDUSTRY LEADERS

To contextualize the FinFET versus GAAFET transition, we examine the strategies adopted by leading foundries at the 3 nm and 2 nm technology nodes. Table 3 compares the architectural choices of TSMC, Samsung, Intel, and IBM, alongside reported improvements in performance, power, and area. Figure 4 illustrates scaling trends in

technology nodes and device density. Figure 5 shows the claimed power reduction at the 3 nm and 2 nm nodes by major manufacturers.

Table 3. Comparison of 3 nm-class technology strategies by company.

Company	Node / Year	Transistor Architecture	Reported Gains (vs previous node)	Status (Year)
TSMC	N3 (~3 nm), 2022 - 23	FinFET (optimized)	+10 - 15% perf or -25 - 30% power vs N5 ¹⁶	Mass production (2023)
Samsung	3 nm (SF3), 2022 - 24	GAAFET (Nanosheet, MBCFET™)	-45% power, +23% perf, -16% area vs 5 nm ¹¹	Initial prod. (2022), volume by 2024
Intel	20A (~2 nm), 2024	GAAFET (RibbonFET)	~+15% energy efficiency vs Intel 4 ¹⁹	Risk production (2024), products ~2025
IBM	2 nm (research), 2021	GAAFET (Nanosheet)	+45% perf or -75% power vs 7 nm	Demo prototype (2021)

TSMC N3 (~3 nm, 2022 - 2023):

TSMC chose to retain FinFETs for its first-generation 3 nm node, branded as N3. By extensively optimizing fin dimensions and fabrication processes, TSMC achieved a reported 10 - 15% improvement in performance at iso-power, or a 25 - 30% reduction in power at iso-performance, relative to its 5 nm (N5) process.²¹ The decision to stay with FinFETs was driven by the ability to meet performance targets without altering the device architecture, thereby leveraging the existing FinFET manufacturing infrastructure and mitigating risks associated with adopting a new device type. However, N3 is widely expected to be TSMC’s final FinFET node for high-performance logic. According to the company’s roadmap, it plans to transition to nanosheet-based transistors at the 2 nm generation (referred to as N2), with a projected rollout around 2025.

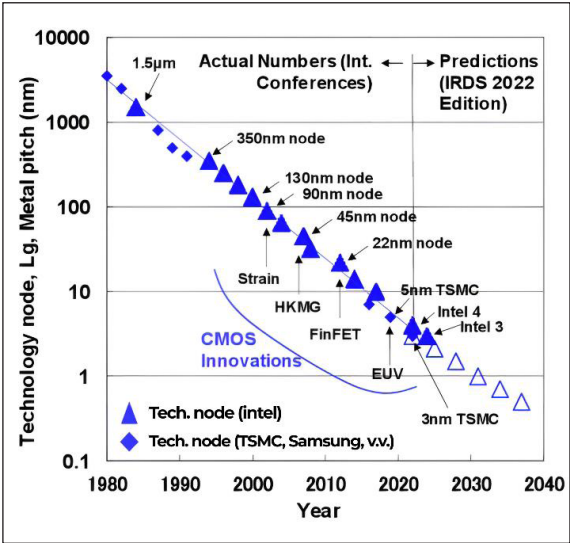


Figure 4. CMOS node shrink from 1.5 μm (1980) to 3 nm (2022) with IRDS-2022 projections toward sub-2 nm. Filled symbols show reported nodes (▲ Intel, ◆ TSMC/Samsung/others); open symbols △ mark IRDS forecasts. Key milestones-strain, HKMG, FinFET, EUV, and GAAFET-are annotated on the log-scale plot of gate length/metal pitch versus year.

Samsung 3 nm (2022 pilot, 2024 volume):

Samsung adopted a more aggressive strategy, becoming the first company in the industry to introduce a GAAFET-based technology into commercial production. At the 3 nm node, Samsung deployed its Multi-Bridge Channel FET (MBCFET™) architecture, based on horizontally stacked silicon nanosheets. Initial chip production on the 3 nm GAA process began in 2022. The company reported substantial improvements over its 5 nm FinFET process, including up to a 45% reduction in power consumption, 23% performance gain, and 16% area reduction.¹³ These gains stem from both architectural advancements and process improvements. By 2024, Samsung aims to scale this technology for high-volume manufacturing and is concurrently developing a second-generation 3 nm GAAFET with anticipated performance gains of up to 30% and power savings of 50% relative to 5 nm. Samsung’s early adoption has allowed it to gain a first-mover advantage in refining GAA

processes, though initial challenges such as yield optimization and ecosystem maturity remain.

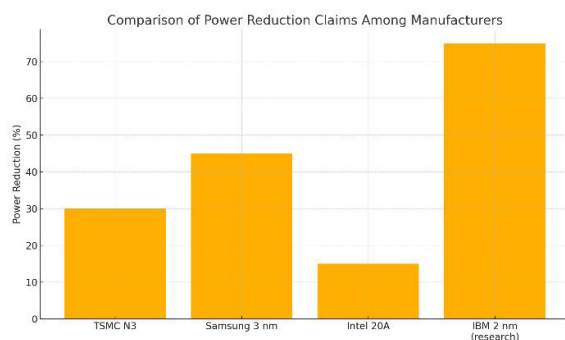


Figure 5. Claimed power reduction for 3 nm and 2 nm nodes by major manufacturers (TSMC, Samsung, Intel, IBM).

Intel 20A (~2 nm, 2024):

Intel plan to introduce its RibbonFET architecture - a proprietary implementation of nanosheet GAAFETs - at the “20A” node (20 Ångströms, approximately equivalent to 2 nm). RibbonFET will be paired with Intel’s backside power delivery network technology, PowerVia.²⁶ According to Intel, RibbonFET is expected to deliver a 15% improvement in energy efficiency compared to Intel 4, either through higher performance at the same power or lower power at the same performance level.²⁷ The architecture involves stacking multiple horizontal nanoribbons for both PMOS and NMOS transistors and is central to Intel’s goal of regaining leadership in process performance. Intel’s 20A timeline aligns with TSMC’s N2 roadmap, with early risk production reported in 2024 and commercial product release targeted for 2025. Intel’s adoption of GAAFET technology reinforces the growing consensus that gate-all-around transistors are essential at the 2 nm node and beyond.²⁸

IBM 2 nm GAAFET prototype (2021):

In 2021, IBM - working in collaboration with research partners - demonstrated a 2 nm test chip fabricated using nanosheet GAAFETs.²⁹

The prototype contained 50 billion transistors on a compact chip footprint and showcased significant performance and efficiency potential: up to 45% performance improvement or 75% power savings relative to a 7 nm baseline.³⁰ While not intended for commercial deployment, the demonstration used a 300 mm wafer fabrication platform and featured gate lengths in the 12 - 14 nm range with vertically stacked nanosheets. IBM’s prototype served as a key proof-of-concept for the feasibility of nanosheet transistors at extremely scaled dimensions, boosting industry confidence in GAAFET adoption.

TSMC’s cautious, infrastructure-leveraged approach and Samsung’s aggressive early GAAFET deployment highlight contrasting risk strategies. Intel’s plan solidifies the view that GAAFETs are essential for scaling to 2 nm. IBM’s research, though exploratory, helped validate the technical path that others are now commercializing. Beyond these four leaders, other foundries such as GlobalFoundries and SMIC are also investigating GAAFETs, though they remain behind in advanced node implementation.

Overall, the semiconductor industry has converged on a clear consensus: GAAFETs are the dominant transistor architecture for the 3 nm and 2 nm nodes. By the mid-2020s, all major vendors are expected to incorporate some form of gate-all-around architecture into their manufacturing processes.

4. POTENTIAL OF 2D MATERIALS FOR POST-3 NM TRANSISTOR SCALING

While silicon-based gate-all-around field-effect transistors (GAAFETs) are expected to sustain Moore’s Law down to approximately 2 nm technology nodes, extending transistor scaling beyond this limit likely necessitates novel channel materials. Among the most promising candidates are two-dimensional (2D) materials - crystalline structures only a few atoms thick - which have garnered substantial attention as

channels for sub-1 nm transistor technologies. Transition metal dichalcogenides (TMDs), such as molybdenum disulfide (MoS_2) and tungsten diselenide (WSe_2), are the most extensively studied representatives of this material class. These materials naturally form layered monolayers ~ 0.7 nm thick, while retaining semiconducting properties, including appropriate bandgaps (~ 1.8 eV for monolayer MoS_2).

A key advantage of monolayer 2D semiconductors lies in their exceptional electrostatic control. Because the channel comprises a single atomic layer, the gate can modulate all carriers directly, eliminating deep-channel effects. This enables ultra-short gate lengths with strong suppression of short-channel effects. For instance, monolayer MoS_2 FETs have demonstrated operation at 1 nm gate lengths using carbon nanotube gates, with acceptable on/off current ratios – performance metrics unachievable with bulk silicon due to leakage currents at such scales.¹⁴

Another intrinsic benefit is the absence of surface dangling bonds. Both surfaces of a monolayer are naturally passivated, which facilitates atomically clean interfaces with gate dielectrics. In contrast to silicon, which requires careful thermal oxidation to minimize interface traps, 2D semiconductors can sustain steep subthreshold swings close to the thermionic limit (~ 60 mV/dec) even at short channel lengths.³¹ High-performance MoS_2 transistors, for example, have demonstrated subthreshold swings around 70 mV/dec, and theoretical studies suggest that values below 60 mV/dec may be achievable using advanced gating methods.³²

Although atomic in thickness, 2D materials exhibit moderate to high carrier mobilities. While silicon mobility declines rapidly at ultra-thin dimensions due to surface roughness and quantum confinement, monolayer MoS_2 devices have exhibited room-temperature electron mobilities of ~ 200 $\text{cm}^2/\text{V}\cdot\text{s}$ in short-

channel configurations.³³ While this is lower than bulk silicon, the robustness of MoS_2 at the atomic scale is significant. Moreover, other 2D materials such as black phosphorus and heterostructured TMDs may offer even higher mobilities. Importantly, 2D semiconductors can often tolerate stronger electrostatic doping and gating fields, providing more flexibility for threshold voltage tuning.

Integration with advanced device architectures

2D semiconductors are being explored not only for planar FETs but also in GAAFET configurations.³⁴ For example, researchers have implemented monolayer MoS_2 and WSe_2 within GAA structures, where the gate completely wraps around the 2D channel encapsulated by insulators. Recent demonstrations have achieved record-high drive currents ($I_{\text{on}} \approx 90$ $\mu\text{A}/\mu\text{m}$) and subthreshold swings as low as 85 mV/dec, even at extremely short gate lengths.³⁵ While these results still trail behind those of silicon-based GAAFETs, improvements are accelerating. Furthermore, the vertical stackability of 2D materials opens pathways toward monolithic 3D integration, including complementary FET (CFET) architectures - such as stacking a p-type WSe_2 FET atop an n-type MoS_2 FET.

Challenges for scalable manufacturing

Despite their promise, significant challenges must be overcome to integrate 2D materials into high-volume semiconductor manufacturing.

Wafer-scale synthesis:

Current approaches for producing high-quality 2D layers - such as mechanical exfoliation or small-scale chemical vapor deposition (CVD) - are inadequate for industrial adoption. For CMOS fabs, it is essential to develop processes capable of growing uniform monolayer films across 300 mm wafers. Techniques like metal-organic CVD (MOCVD) have shown promise for MoS_2 and WS_2 , but achieving high monolayer uniformity ($>99\%$)

and low defect densities ($<10^8 \text{ cm}^{-2}$) remains a major challenge.³² Advanced approaches such as seeded growth, phase-selective deposition, and surface functionalization are being investigated to meet these stringent requirements.¹⁴

Contact resistance:

Forming low-resistance ohmic contacts to 2D semiconductors is particularly challenging due to the absence of out-of-plane bonds and Fermi level pinning, which lead to high Schottky barriers and contact resistances ranging from tens to hundreds of $\text{k}\Omega\cdot\mu\text{m}$.¹⁴ One promising approach involves phase engineering, where the semiconducting 2H phase of MoS_2 is locally converted to a metallic 1T' phase, improving band alignment and reducing resistance.³¹ Other strategies include edge-contact geometries, where electrodes contact the exposed edges of 2D sheets, and the insertion of interfacial layers such as graphene or metallic TMDs. Some configurations have achieved $<1 \text{ k}\Omega\cdot\mu\text{m}$ contact resistance, though further innovation is required to match silicon's performance ($\sim 10 - 50 \Omega\cdot\mu\text{m}$).^{34,35}

Dielectric integration and thermal stability:

2D semiconductors must be compatible with high- κ gate dielectrics like HfO_2 and withstand backend-of-line (BEOL) thermal budgets ($\sim 400^\circ\text{C}$). Although their dangling-bond-free surfaces enable low interface trap densities, defects can form during dielectric deposition, especially with plasma-assisted atomic layer deposition (ALD). Researchers are exploring gentler deposition methods, including surface pre-functionalization to initiate uniform nucleation without channel damage. Ensuring long-term stability in mobility and threshold voltage under thermal and electrical stress remains a critical area of investigation.³¹

2D semiconductors represent a compelling solution for extending transistor scaling into the angstrom regime, potentially mitigating the electrostatic and physical limitations of bulk

silicon. While integration into mainstream CMOS requires substantial advances in materials science, device engineering, and process technology, progress is steady. Each year brings improvements in device performance metrics -including current drive, subthreshold swing, and contact resistance – as well as better methods for large-area monolayer synthesis. Within the next 5-10 years, it is plausible that 2D FETs may be deployed in niche low-power applications or 3D-stacked logic architectures. Section 5 will examine how these advances align with broader fabrication and system-level integration challenges.

5. FABRICATION CHALLENGES AND HETEROGENEOUS INTEGRATION STRATEGIES

As semiconductor scaling advances toward the 3 nm, 2 nm, and sub-2 nm nodes, fabrication challenges have become as significant as innovations in device architecture. This section highlights key manufacturing obstacles at these advanced nodes and explores heterogeneous integration strategies that complement traditional scaling.

EUV lithography limitations

The introduction of extreme ultraviolet (EUV) lithography at the $\sim 7 \text{ nm}$ node has enabled further miniaturization. However, despite operating at a 13.5 nm wavelength, EUV systems face fundamental resolution limits for features below 3 nm. While high-numerical-aperture (high-NA) EUV is expected to extend patterning capabilities, sub-3 nm features often still require multiple-patterning techniques to define nanosheets, fins, and tightly spaced interconnects.

Each additional patterning step increases complexity, variability, and cost. Moreover, stochastic effects inherent to EUV-such as photon shot noise and resist bridging-introduce random defects at the nanometer scale. As a result, the industry is investing in resist development, anti-

collapse formulations, and restrictive design rules to maintain pattern fidelity. The deployment of EUV and multi-patterning technologies has also led to a sharp rise in wafer costs, making the economic feasibility of advanced nodes a critical concern.

Process variability and control

At these dimensions, atomic-scale control is essential. In gate-all-around FETs (GAAFETs), variations in nanosheet or gate dielectric thickness across the wafer on the order of fractions of a nanometer can cause substantial shifts in threshold voltage and leakage current. Ma et al.²⁴ specifically identified that even minor variations in source/drain height substantially affect device characteristics, underscoring the critical need for rigorous atomic-scale process control at sub-3 nm nodes. Additionally, Zhu et al.²⁵ have shown that stress-induced mobility enhancement through Si/SiGe hybrid-channel integration can yield substantial performance gains, further highlighting strain engineering as crucial for sub-3 nm transistor optimisation. To meet these tolerances, advanced process techniques such as atomic layer deposition (ALD) and atomic layer etching (ALE) are employed.

Equally critical is metrology: in-line measurement of sub-nanometer features is non-trivial. Tools like spectroscopic ellipsometry, CD-SEM, and cross-sectional TEM are often used, but typically limited to test structures. Furthermore, classical ion implantation is being replaced due to variability at small scales. Instead, threshold tuning is achieved via in-situ doped epitaxial source/drain regions or through work-function modulation of gate metals.

Thermal and Power Density Constraints

Even though individual transistors become more energy-efficient, increasing transistor density drives up overall power density. Thermal management becomes more challenging as billions of transistors are densely integrated on a

single die. Power delivery architecture is critical to mitigating thermal hotspots and IR drop.

Intel's PowerVia architecture exemplifies innovation in this area. By delivering power from the backside of the wafer in conjunction with RibbonFET technology, PowerVia improves IR drop, reduces front-side routing congestion, and enhances thermal uniformity. Nevertheless, design constraints such as "dark silicon" - where not all transistors can be activated simultaneously due to thermal limitations - remain a concern.

Cost and yield considerations

Historically, cost per transistor declined with each successive node. However, at 5 nm and beyond, this trend has slowed or even reversed.¹⁵ EUV lithography, increased process complexity, and reduced yield have significantly raised wafer costs, particularly at 3 nm. These cost pressures are driving a paradigm shift toward chiplet-based architectures.

Rather than producing a single monolithic die at the latest node, designers can partition functionality into smaller chiplets. Critical logic blocks can be fabricated at 3 nm, while less performance-sensitive blocks such as I/O and analog functions are implemented on mature nodes like 14 nm. These chiplets are then integrated via advanced packaging techniques, optimizing overall performance, yield, and cost.¹³

3D heterogeneous integration and emerging architectures

Beyond 2.5D chiplet integration, three-dimensional (3D) stacking is emerging as a key strategy for density scaling. Through-silicon vias (TSVs) and wafer-to-wafer bonding enable vertical stacking of dies, such as memory-on-logic or logic-on-logic configurations.

Of particular interest is the Complementary FET (CFET) architecture, where NMOS and PMOS transistors are stacked vertically rather than placed laterally.²³ This configuration doubles

effective transistor density while maintaining CMOS functionality. CFETs can be realized by vertically stacking two GAAFETs using either silicon nanosheets or 2D materials. While early demonstrations confirm CFET feasibility, precise alignment and vertical interconnect formation remain formidable challenges.

Another novel approach is the Vertical Transport FET (VTFET), pioneered by IBM and others. In VTFETs, the current flows vertically from source to drain, and the gate wraps around a vertical channel. This configuration offers over $2\times$ density improvements compared to planar transistors, with potential performance gains.¹⁷

Both CFET and VTFET architectures are still in the research and development phase but are being actively explored as potential successors to GAAFETs when lateral scaling saturates.

Design-technology co-optimization (dtco) and system-level gains

The increasing complexity of advanced nodes necessitates design-technology co-optimization (DTCO). Designers must understand manufacturing constraints, while process engineers must provide levers (e.g., new devices, layout options) that designers can exploit. DTCO is critical to extracting maximum value from each technology generation.

Heterogeneous integration is increasingly viewed as the third pillar of progress, alongside device scaling and circuit-level innovation. System-level benefits such as reduced memory access latency and lower energy per operation can be achieved by vertically stacking memory and logic. Similarly, chiplet designs enable function-specific optimization across different process nodes, improving both performance-per-watt and performance-per-dollar.

Scaling to 3 nm and beyond is no longer just a matter of shrinking transistors. It requires multidisciplinary innovation across materials science (e.g., novel channels and dielectrics),

electrical engineering (e.g., CFETs, VTFETs), and computer architecture (e.g., chiplet systems, DTCO). Heterogeneous integration is now a cornerstone strategy for sustaining Moore's Law-allowing meaningful system-level advancements even when traditional transistor scaling slows. The next section will explore the broader implications of these developments on the future of semiconductor technology.

6. CONCLUSION AND OUTLOOK

Transistor scaling has reached a stage where geometric and material innovations are as essential as dimensional reduction. Gate-All-Around FETs (GAAFETs), notably nanosheet transistors, have demonstrated their viability at the 3 nm node, delivering improved performance and energy efficiency. This analysis confirms that GAAFETs outperform FinFETs in drive current, electrostatic control, and leakage, validating the industry's shift to GAAFETs for 3 nm and 2 nm technologies. Early deployments by Samsung and Intel further affirm GAAFETs as practical solutions for next-generation CMOS.

Looking ahead, several critical directions are emerging:

- *New materials for scaled channels:* Two-dimensional (2D) semiconductors offer compelling properties-atomic-scale thickness, strong gate control, and reduced short-channel effects-that make them prime candidates for sub-2 nm technologies. Hybrid stacks, such as CFETs combining a silicon GAAFET and a 2D FET, may become a key architecture. High-mobility III-V compounds and strain-engineered SiGe are also under exploration for performance-critical applications like RF or analog blocks.

- *Beyond-GAAFET device architectures:* Vertical transport FETs (VTFETs) and Complementary FETs (CFETs) extend scaling into the third dimension. VTFETs rotate the transistor vertically to improve density; CFETs vertically stack NMOS and PMOS to halve area. Lab demonstrations show promise,¹⁷ but

challenges in manufacturability and circuit reliability remain. These devices could succeed GAAFETs later this decade.

• *Heterogeneous integration and system co-design*: As transistor pitch scaling slows, progress continues through packaging and architectural co-design-the essence of “More-than-Moore.” Future systems will leverage 2.5D and 3D integration, combining chiplets across nodes and technologies (logic, memory, photonics) within a single package. Success demands cross-disciplinary collaboration spanning materials, devices, circuits, and systems.

In summary, the sub-3 nm roadmap presents formidable challenges, but also vast potential. GAAFETs have opened a path forward for near-term scaling, while emerging materials and 3D device architectures promise long-term extension. Realizing this future will require synergy between device physics, fabrication technology, and system design. With continued collaboration between academia and industry, the momentum of Moore’s Law can be sustained-powering breakthroughs in computing, communications, and beyond.

Acknowledgment

The author expresses sincere thanks to colleagues and mentors who provided feedback and support during the research. No external funding was used in this study.

REFERENCES

1. M. T. Bohr, I. A. Young. CMOS scaling trends and beyond, *IEEE Micro*, **2017**, 37, 20 - 29.
2. S. K. Saha. *FinFET devices for VLSI circuits and systems*, CRC Press, Boca Raton, 2020.
3. C. H. Lin, B. Greene, S. Narasimha, J. Cai, A. Bryant, C. Radens, V. Narayanan, B. Linder, H. Ho, A. Aiyar, et al. High performance 14 nm SOI FinFET CMOS technology with 0.0174 μm^2 embedded DRAM and 15 levels of Cu metallization, *IEEE International Electron Devices Meeting*, **2014**, 381-383.
4. S. Y. Wu, C. Y. Lin, M. C. Chiang, J. J. Liaw, J. Y. Cheng, S. H. Yang, M. Liang, T. Miyashita, C. H. Tsai, B. C. Hsu, et al. A 16 nm FinFET CMOS technology for mobile SoC and computing applications, *2013 IEEE International Electron Devices Meeting*, **2013**, 911-914.
5. K. Karimi, A. Fardoost, M. Javanmard. Comprehensive review of FinFET technology: history, structure, challenges, innovations, and emerging sensing applications, *Micromachines*, **2024**, 15, 1187.
6. H. Wong, K. Kakushima. On the vertically stacked gate-all-around nanosheet and nanowire transistor scaling beyond the 5 nm technology node, *Nanomaterials*, **2022**, 12, 1748.
7. U. Soma. Transistor evolution: a comprehensive overview from TFT to TFET and beyond, *Proceedings of the National Academy of Sciences, India Section A: Physical Sciences*, **2025**, 95, 113-125.
8. R. R. Das, T. R. Rajalekshmi, A. James. FinFET to GAA MBCFET: a review and insights, *IEEE Access*, **2024**, 12, 50556-50577.
9. W. Hu, F. Li. *Scaling beyond 7 nm node: an overview of gate-all-around FETs*, 2021 9th International Symposium on Next Generation Electronics (ISNE), Changsha, China, 2021.
10. J. S. Yoon, J. Jeong, S. Lee, R. Baek. Optimization of nanosheet number and width of multi-stacked nanosheet FETs for sub-7-nm node system on chip applications, *Japanese Journal of Applied Physics*, **2019**, 58, 084505.
11. S. Kim, K. Lee, S. Kim, M. Kim, J. H. Lee, S. Kim, B. G. Park. Investigation of device performance for fin angle optimization in FinFET and gate-all-around FETs for 3 nm-node and beyond, *IEEE Transaction Electron Devices*, **2022**, 69, 2088-2093.

12. S. Barraud, B. Previtali, C. Vizioz, J. M. Hartmann, J. Sturm, J. Lassarre, C. Perrot, P. Rodriguez, V. Loup, A. M. Lucas, et al. *7-Levels-stacked nanosheet GAA transistors for high performance computing*, 2020 IEEE Symposium on VLSI Technology, Honolulu, HI, USA, 2020.
13. J. Jeong, S. H. Lee, S. A. Masuoka, S. Min, S. Lee, S. Kim, T. Myung, B. Choi, C. W. Sohn, S. W. Kim, et al. *World's first GAA 3 nm foundry platform technology (SF3) with novel multi-bridge-channel-FET (MBCFETTM) process*, 2023 IEEE Symposium on VLSI Technology and Circuits, Kyoto, Japan, 2023.
14. K. Maxey, C. H. Naylor, K. P. O'Brien, A. Penumatcha, A. Oni, C. Mokhtarzadeh, C. J. Dorow, C. Rogan, B. Holybee, T. Tronic, et al. *300 mm MOCVD 2D CMOS materials for more (than) moore scaling*, 2022 IEEE Symposium on VLSI Technology and Circuits, Honolulu, HI, USA, 2022.
15. S. Chen, H. Zhang, Z. Ling, J. Zhai, B. Yu. *The survey of 2.5D integrated architecture: an EDA perspective*, Proceedings of the 30th Asia and South Pacific Design Automation Conference, Association for Computing Machinery, New York, USA, 2025.
16. S. Joung, S. Kim. *Leakage performance improvement in multi-bridge-channel field effect transistor (MBCFET) by adding core insulator layer*, 2019 International Conference on Simulation of Semiconductor Processes and Devices (SISPAD), IEEE, Udine, Italy, 2019.
17. D. E. Shim, P. Kumar, A. A. Kini, M. Mallikarjuna, M. N. H. Shazon, A. Naeemi. *GT3: an open-source 3 nm GAAFET PDK and platform for end-to-end evaluation of emerging technologies*, *IEEE Transactions on Electron Devices*, **2025**, 72, 1582-1588.
18. L. Scognamiglio, F. Mo, C. E. Spano, M. Vacca, G. Piccinini. *Simulation study on the impact of miniaturization in 3 nm node 3D junctionless transistors*, *IEEE Transactions on Nanotechnology*, **2025**, 24, 102-109.
19. N. Totorica, W. Hu, F. Li. *Simulation of different structured gate-all-around FETs for 2 nm node*, *Engineering Research Express*, **2024**, 6(3), 1-11.
20. L. Abdelaziz, B. Khaled, G. Mustapha. *Comparative analysis of TG FinFET and GAA FinFET in 3 nm technology node*, 2023 27th International Conference on Information Technology (IT), Zabljak, Montenegro, 2023.
21. S. Y. Wu, C. H. Chang, M. C. Chiang, C. Y. Lin, J. J. Liaw, J. Y. Cheng, J. Y. Yeh, H. F. Chen, S. Y. Chang, K. T. Lai, et al. *A 3 nm CMOS FinFlexTM platform technology with enhanced power efficiency and performance for mobile SoC and high performance computing applications*, 2022 International Electron Devices Meeting (IEDM), IEEE, San Francisco, CA, USA, 2022, 2751-2754.
22. K. Saini, V. Kumar. *Comparative analysis of 12 nm gate-all-around vertically stacked nanosheet field effect transistor for high-k gate-oxide dielectrics*, 2024 First International Conference on Electronics, Communication and Signal Processing (ICECSP), New Delhi, India, 2024.
23. D. Wang, X. Sun, T. Liu, K. Chen, J. Yang, C. Wu, M. Xu, W. Zhang. *Investigation of source/drain recess engineering and its impacts on FinFET and GAA nanosheet FET at 5 nm node*, *Electronics*, **2023**, 12, 0770.
24. M. Ma, C. Li, J. Ma, W. Yang, H. Li, H. You, M. J. Deen. *Investigation of source/drain height variation and its impacts on FinFET and GAA nanosheet FET*, *Electronics*, **2025**, 14, 1091.
25. C. Zhu, X. Zhu, S. Yu, D. Wei Zhang. *Process-dependent evolution of channel stress and stress-induced mobility gain in FinFET, normal GAAFET, and Si/SiGe hybrid channel GAAFET*, *IEEE Access*, **2025**, 13, 21600-21609.
26. X. Wang, Y. Kim, G.H. Baek, K.G. Bannore, K. Dave, A. Joushaghani, N. Kang, M. Ko, A.M. Pillai, H.C. Prakash Movva, G. Park, M. Rahman, S. Subramaniam, V. Vashishtha,

- T. Yang, Z. Guo, E. Karl. *A 0.021 μm^2 high-density SRAM in Intel-18A-RibbonFET technology with powervia-backside power delivery*, 2025 IEEE International Solid-State Circuits Conference (ISSCC), IEEE, San Francisco, CA, USA, 2025.
27. A. Agrawal, W. Chakraborty, W. Li, H. Ryu, B. Markman, S. H. Hoon, R. K. Paul, C. Y. Huang, S. M. Choi, K. Rho, et al. *Silicon ribbon FET CMOS at 6 nm gate length*, 2024 IEEE International Electron Devices Meeting (IEDM), San Francisco, CA, USA, 2024.
 28. W. Hafez, P. Agnihotri, M. Asoro, M. Aykol, B. Bains, R. Bambery, M. Bapna, A. Barik, A. Chatterjee, P. C. Chiu, et al. *Intel powervia technology: backside power delivery for high density and high-performance computing*, 2023 IEEE Symposium on VLSI Technology and Circuits (VLSI Technology and Circuits), Kyoto, Japan, 2023.
 29. Y. Chen, V. Sankatali, S. Mishra, J. Ryckaert, J. Myers, D. Biswas. *Thermal implications in scaling high-performance server 3D chiplet-based 2.5D SoC from FinFET to nanosheet*, 2024 IEEE Computer Society Annual Symposium on VLSI (ISVLSI), Knoxville, TN, USA, 2024.
 30. N. Loubet, T. Hook, P. Montanini, C. W. Yeung, S. Kanakasabapathy, M. Guillom, T. Yamashita, J. Zhang, X. Miao, J. Wang, et al. *Stacked nanosheet gate-all-around transistor to enable scaling beyond FinFET*, 2017 Symposium on VLSI Technology, Kyoto, Japan, 2017.
 31. T. Knobloch, B. Uzlu, Y. Yu. Illarionov, Z. Wang, M. Otto, L. Filipovic, M. Walzl, D. Neumaier, M. C. Lemme, T. Grasser. *Improving stability in two-dimensional transistors with amorphous gate oxides by Fermi-level tuning*, *Nature Electronics*, **2022**, 5, 356-366.
 32. P. C. Shen, C. Su, Y. Lin, A. S. Chou, C. C. Cheng, J. H. Park, M. H. Chiu, A. Y. Lu, H. L. Tang, M. M. Tavakoli, et al. *Ultralow contact resistance between semimetal and monolayer semiconductors*, *Nature*, **2021**, 593, 211-217.
 33. K. Kwak, H. Yoon, S. Hong, B. H. Kang. *Advances in 2D molybdenum disulfide transistors for flexible and wearable electronics*, *Micromachines*, **2024**, 15, 1476.
 34. B. J. Chou, Y. Y. Chung, W. S. Yun, C. F. Hsu, M. Y. Li, S. K. Su, S. L. Liew, V. D. H. Hou, C. W. Chen, C. C. Kei, et al. *High-performance monolayer MoS_2 nanosheet GAA transistor*, *Nanotechnology*, **2024**, 35, 125204.
 35. W. Mortelmans, P. Buragohain, C. Rogan, A. Kitamura, C. J. Dorow, K. P. O'Brien, R. Ramamurthy, J. Lux, T. Zhong, S. Harlson, et al. *Record performance in GAA 2D NMOS and PMOS using monolayer MoS_2 and WSe_2 with scaled contact and gate length*, 2024 IEEE Symposium on VLSI Technology and Circuits (VLSI Technology and Circuits), Honolulu, HI, USA, 2024.



© 2025 by the authors. This Open Access Article is licensed under the Creative Commons Attribution-NonCommercial 4.0 International (CC BY-NC 4.0) license (<https://creativecommons.org/licenses/by-nc/4.0/>).

Tổng hợp vật liệu Co_3O_4 có nguồn gốc từ ZIF-67 và ứng dụng trong biến tính điện cực phát hiện acid ascorbic

Ngô Thị Thanh Hiền*, Nguyễn Vũ Ngọc Mai

Khoa Khoa học Tự nhiên, Trường Đại học Quy Nhơn, Việt Nam

Ngày nhận bài: 09/04/2025; Ngày sửa bài: 25/06/2025;
Ngày nhận đăng: 18/07/2025; Ngày xuất bản: 28/10/2025

TÓM TẮT

Trong nghiên cứu này, Co_3O_4 dạng tinh thể có cấu trúc xếp được tổng hợp bằng cách nung ZIF-67 trong môi trường không khí. Đặc trưng vật liệu Co_3O_4 được nghiên cứu bằng XRD, BET, SEM và EDS. Điện cực biến tính Co_3O_4 -GPE dùng phát hiện điện hóa acid ascorbic thể hiện khoảng tuyến tính từ 2 μM đến 15 μM với giới hạn phát hiện là 0,48 μM . Kết quả độ thu hồi dao động từ 97,82% đến 99,5% đối với acid ascorbic xác định trong viên thuốc thương mại.

Từ khóa: Co_3O_4 , ZIF-67, acid ascorbic.

*Tác giả liên hệ chính.

Email: ngothanhhien@qnu.edu.vn

ZIF-67-derived Co_3O_4 porous crystalline material for electrochemical ascorbic acid detection

Ngo Thi Thanh Hien*, Nguyen Vu Ngoc Mai

Faculty of Natural Sciences, Quy Nhon University, Vietnam

Received: 09/04/2025; Revised: 25/06/2025;

Accepted: 18/07/2025; Published: 28/10/2025

ABSTRACT

The Co_3O_4 porous crystalline material was synthesized by calcining ZIF-67 sample in air. The Co_3O_4 material was characterized by XRD, BET, SEM, and EDS. The electrode modified with Co_3O_4 was used to determine ascorbic acid. The proposed Co_3O_4 -GPE electrode exhibited a linear range of 2 μM to 15 μM with a detection limit of 0.48 μM . Recovery results, ranging from 97.82% to 99.5% for ascorbic acid in pharmaceutical tablet.

Keywords: Co_3O_4 , ZIF-67, ascorbic acid.

1. INTRODUCTION

Ascorbic acid (AA), the common name for Vitamin C, is a common multivitamin component and occurs naturally in various foods. It is important for a healthy diet and acts as an antioxidant. However, an overdose of vitamin C can lead to side effects such as stomach upset, headache, difficulty sleeping, and skin flushing.^{1,2} Therefore, the rapid and accurate determination of AA has attracted scientific attention.

Many analytical methods exist for determining ascorbic acid (AA), including techniques such as spectrofluorometry,^{3,4} chromatography,^{5,6} spectrophotometry,^{7,8} capillary zone electrophoresis,^{9,10} and electrochemistry.^{11,12} Among these, electrochemical methods employing modified electrodes have received considerable interest owing to their inherent simplicity, high sensitivity, and economic viability.

Cobalt oxide is a semiconductor with wide applications in many fields, including catalysis,

electrode materials, gas sensing, and drug delivery.¹³⁻¹⁵ Numerous studies have explored the diverse applications of Co_3O_4 ; however, its potential use in electrode modification for pharmaceutical analysis remains relatively underexplored. To date, various porous nanostructures of Co_3O_4 have been synthesized, including spherical, tubular, rod-like, and flower-like morphologies. Most synthesis methods utilize cobalt carbonate or hydroxide salts as precursors, often yielding materials with relatively low surface areas.^{16,17}

Recent, the application of metal-organic frameworks (MOFs; ZIFs) as precursors in the synthesis of inorganic materials is a growing area of research.¹⁸⁻²⁵ Studies show that heat treatment of ZIF-67 can pyrolyze their ligands and lead to the formation of metal oxide nanoparticles. Therefore, the metal-centered organic framework material Co (ZIF-67) has appeared as a potential precursor to synthesize cobalt oxide (Co_3O_4) while still inheriting the structural characteristics of ZIF-67 and improving its catalytic activity.

*Corresponding author.

Email: ngothanhhien@qnu.edu.vn

In this work, an electrode modified with the Co_3O_4 porous crystalline material derived from ZIF-67 is investigated. The obtained electrode was used for the electrochemical determination of AA.

2. EXPERIMENT

2.1. Chemicals

2-methylimidazol (2-H_{mim} , 98%), ascorbic acid, graphite powder and paraffin oil were received from Sigma Aldrich. Cobalt nitrate hexahydrate [$\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, 99%] was purchased from Macklin (China). Phosphoric acid (H_3PO_4 , 85%), potassium dihydrogen phosphate (KH_2PO_4 , 99%), boric acid (H_3BO_3 , 99%) and potassium hydroxide (KOH) were received from Guangdong-Guanghua Co. Ltd (China).

Vitamin C tablet (Vitamin C, 500 mg AA, from Pharimexco Viet Nam) was purchased from a local pharmacy. All chemical reagents were used as received without any further purification.

Britton-Robinson (B-R) buffer solutions were made using 0.5 M solutions of H_3BO_3 , H_3PO_4 , and CH_3COOH . The pH of the B-R buffer was adjusted to the desired value with 1 M KOH or 1M H_3PO_4 solutions.

2.2. Apparatus

All electrochemical analyses, including cyclic voltammetry and square wave voltammetry, were conducted using a DY2322 potentiostat, Digi-Ivy, Inc. Austin. A standard three-electrode cell was employed, consisting of a working electrode (Co_3O_4 -GPE or bare GPE, 0.07 cm^2), a counter electrode (Pt wire), and a reference electrode (Ag/AgCl , KCl_{sat}).

2.3. Synthesis of Co_3O_4 porous crystalline material from ZIF-67

The synthesis of ZIF-67 was conducted according to a previously established method.²⁶

1.455g of $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ was dissolved in 50 mL of ethanol, and 1.64g of 2-methylimidazole (H_{mim}) was dissolved in 50mL of ethanol, resulting in a $\text{Co}^{2+}:\text{H}_{\text{mim}}$ molar

ratio of 1 : 4. The H_{mim} solution was slowly added to $\text{Co}(\text{NO}_3)_2$ solution under continuous stirring for 30 minutes at room temperature. The obtained mixture was allowed to stand at room temperature for 6 hours without stirring, leading to the formation of a colloidal dispersion. The solid product was collected by centrifugation (4000 rpm, 30 minutes), washed three times with ethanol, and dried at $80\text{ }^\circ\text{C}$, 12 hours.

The Co_3O_4 was obtained by calcining ZIF-67 in air at a heating rate of $1\text{ }^\circ\text{C}\cdot\text{min}^{-1}$.

2.4. Preparation of Co_3O_4 -GPE modified electrode

To prepare the Co_3O_4 -GPE modified electrode, 40 mg of graphite powder and 5 mg of Co_3O_4 powder were thoroughly mixed with 10 μL of paraffin oil. The resulting paste was then packed into a Teflon holder, and its surface was smoothed using paper. To renew the electrode surface, the outer 2 mm of paste was removed and replaced with freshly prepared paste.

2.5. Characterization of the Co_3O_4 porous crystalline material

A Bruker-Axs D8 diffractometer (40 kV, 40 mA) was used for powder XRD analysis. Textural properties were determined from nitrogen adsorption-desorption isotherms at $-196\text{ }^\circ\text{C}$ with a Micromeritics Gemini VII 2390 V1.02. Sample morphology was examined by scanning electron microscopy (JEOL JSM-6700F, 15 kV, 10 mA), and elemental composition was analyzed using EDS with a JSM-5700 LV.

3. RESULTS AND DISCUSSION

3.1. Characterization of the synthesized Co_3O_4 material

The XRD diffraction pattern of Co_3O_4 sample are shown in Fig. 1. The X-ray diffraction pattern exhibited reflections at 2θ values of approximately 31.5° , 36.8° , 38.0° , 44.6° , 55.8° , 59.4° , 65.3° and 77.5° , corresponding to the (220), (311), (222), (400), (422), (511), (440) and (533) crystalline planes of the Co_3O_4 cubic structure (JCPDS No. 04-043-1003).²⁵

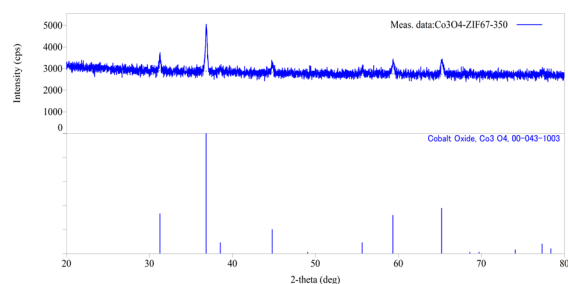


Figure 1. XRD pattern of the Co_3O_4 porous crystalline material.

The nitrogen adsorption-desorption isotherms were employed to characterize the specific surface area and pore morphology of the Co_3O_4 sample. As depicted in Figure 2, the sample presented a Type IV isotherm, accompanied by an H3 hysteresis loop, suggesting a mesoporous structure. The BET surface area was determined to be $30.43 \text{ m}^2/\text{g}$, and the pore size distribution was at 3 nm.

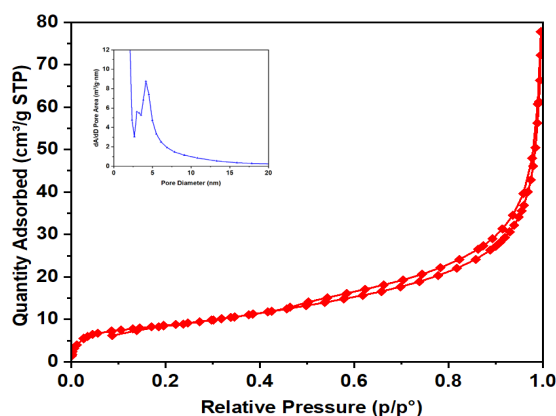


Figure 2. Nitrogen adsorption-desorption isotherms (A) and pore size distribution (B) of the Co_3O_4 porous crystalline material.

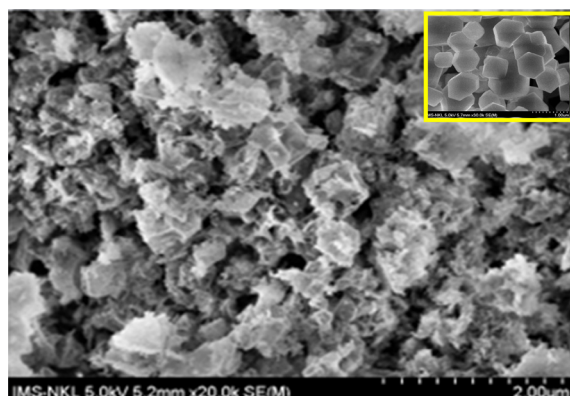


Figure 3. SEM image of the Co_3O_4 porous crystalline material, Inset: SEM image of ZIF-67 material.

The SEM images of Co_3O_4 (Figure 3) revealed that the calcined particles retained a cubic morphology, consistent with the original ZIF-67 crystal template, characterized by an internal hollow structure and a surface exhibiting porosity. However, thermal treatment resulted in the observation of some collapsed hollow structures.

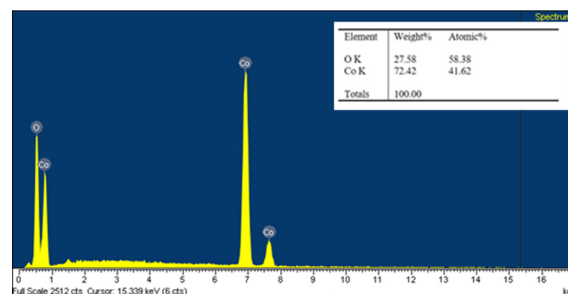


Figure 4. EDS spectra of the Co_3O_4 porous crystalline material.

The EDS analysis was conducted on the Co_3O_4 sample (Figure 4). Cobalt (Co) and oxygen (O) were confirmed to be present on the sample's surface based on the results. The elemental analysis of the Co_3O_4 yielded 41.62% cobalt and 58.38% oxygen.

A comprehensive morpho-structural analysis of the Co_3O_4 material, utilizing results of XRD, SEM, EDS and BET, validated the successful synthesis of the Co_3O_4 porous crystalline material.

3.2. Electrochemical characterization

The electrochemical behavior of ascorbic acid (AA) was investigated using cyclic voltammetry (CV) and square wave voltammetry (SWV).

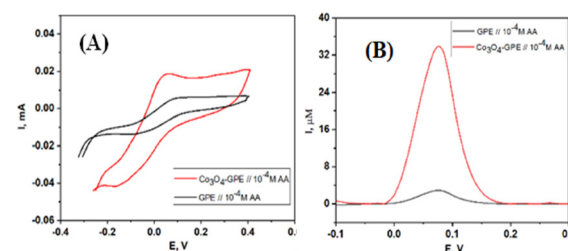


Figure 5. Cyclic voltammograms (A) and square wave voltammograms (B) at GPE and Co_3O_4 -GPE modified electrode in 0.2 M B-R buffer solution pH = 4 containing of 10^{-4} M AA.

A peak of AA at 0.08 V was observed in the CV and SWV curves obtained at both the bare GPE and the Co₃O₄-GPE, as illustrated in Figure 5. The Co₃O₄-GPE exhibited a lower peak potential and higher current. The oxidation peak current for AA at the Co₃O₄-GPE was approximately fifteen-fold greater than that observed at the bare GPE (Figure 5B).

The reaction mechanism of ascorbic acid onto the Co₃O₄ nanostructure takes place by the transfer of two electrons and protons, as shown in Figure 6, and it has been generally represented in the literature.^{27, 28}

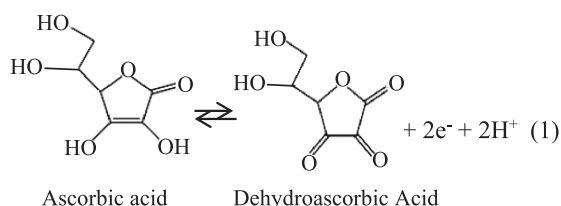


Figure 6. The electro-oxidation reaction of ascorbic acid.

Co₃O₄ showed catalytic activity for ascorbic acid electro-oxidation due to its unique redox properties, containing both Co²⁺ and Co³⁺ states. The catalytic process involves an electrochemical mechanism: surface Co³⁺ is electrochemically oxidized to a highly reactive Co⁴⁺, which then chemically oxidizes ascorbic acid back to dehydroascorbic acid, regenerating Co³⁺. This continuous regeneration ensures sustained catalytic activity. The porosity of Co₃O₄ derived from ZIF-67 further enhances its electrocatalytic performance. Its porous structure provides a large surface area with numerous active sites and facilitates efficient mass transport. This structure also improves the electrode-electrolyte interface and prevents nanoparticle agglomeration, significantly increasing the overall reaction rate and electrode efficiency.^{29,30}

3.3. The effect of pH

The square wave voltammetry (SWV) was employed to examine the impact of pH (within the range of 3 to 6) on the voltammetric signals of AA. The pH of the electrolyte significantly affects the AA oxidation on the modified electrode. Figure 7 displays the current responses

recorded on the Co₃O₄-GPE under different pH conditions.

A substantial increase in peak current was observed as t_{acc} increased from 0 to 90 seconds, suggesting a corresponding enhancement of AA accumulation at the electrode surface. Beyond 90 seconds, however, the peak current exhibited negligible increase, indicative of the electrode surface approaching adsorption equilibrium. Based on this observation, 90 seconds was selected as the optimal t_{acc} .

3.4. Accumulation

The effect of accumulation time (t_{acc}) on electrode response was investigated across a range of 0 to 150 seconds in a 0.2 M B-R buffer (pH 4) with 10⁻⁴ M AA (Figure 8).

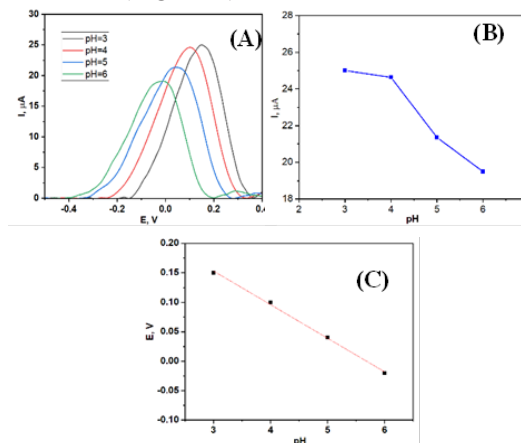


Figure 7. Square wave voltammograms of Co₃O₄-GPE in 0.2 M B-R buffer (pH 4) containing 10⁻⁴ M AA (A); Influence of pH on I_p (B); Plot of E_p vs. pH (C).

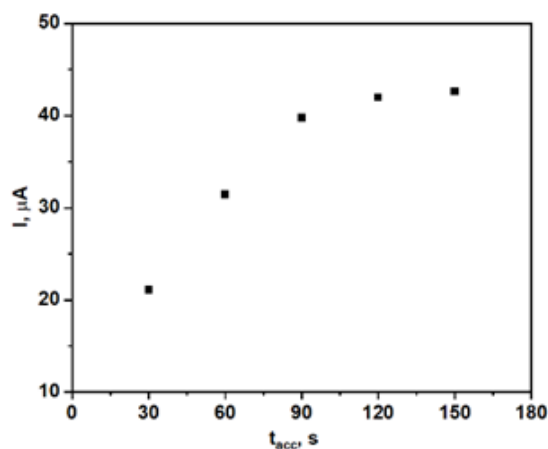


Figure 8. Dependence of I_p for AA in 0.2M B-R buffer solution pH 4 on accumulation time.

3.5. Calibration

The calibration curves for the AA detection with varying concentrations of AA was constructed by recording SWV in 0.2 M B-R buffer solution at pH = 4 (Figure 9A). Accordingly, a calibration curve was shown in Figure 9B. The electrode exhibited a linear response for AA concentrations between 2 μ M and 15 μ M.

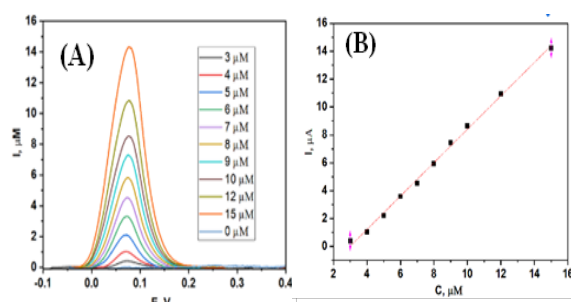


Figure 9. Square wave voltammograms recorded at Co_3O_4 -GPE increasing concentration of AA (A) and the corresponding calibration curve (B).

The resulting linear regression equation was:

$$I_{pa}/\mu\text{A} = (-3.56335 \pm 0.19192) + (1.19706 \pm 0.02218) [\text{AA}]/\mu\text{M}, (R = 0.99726)$$

For ascorbic acid (AA) detection, the Co_3O_4 -GPE electrode exhibited a 0.48 μ M detection limit and a sensitivity of 1.19, which are lower compared to some published results (Table 1).

The continuous regeneration of the $\text{Co}^{2+}/\text{Co}^{3+}$ redox couple in the investigated electrode increases the reaction rate, leading a larger current signal due to enhanced electron transfer. The resulting improved signal directly increases the electrode's sensitivity, allowing for the detection of even minor changes in analyte concentration. Consequently, the limit of detection (LOD) decreases, as a superior signal-to-noise ratio facilitates the reliable detection of very low analyte concentrations.

Table 1. Summary of electrode performance in this work and literature.

Electrode	Method	Detection limit, μ M	References
CL-TiN/GCE	DPV	1.52	31
NiCoO_2/C	Amperometry	0.5	32
AgNP-Psi	Amperometry	0.83	33
GO-XDA- Mn_2O_3	DPV	0.6	34
rGO/Au/GCE	DPV	0.51	35
Co_3O_4 -GPE	SWV	0.48	This work

3.6. Long-term stability and reproducibility

To assess the long-term stability, the electrode was stored at 4 $^{\circ}\text{C}$ in a refrigerator for 7 days, and the peak oxidation currents of 5 μ M AA in a 0.2 M B-R buffer solution were recorded. Measurements were conducted at various times. After 7 days of storage, the peak oxidation current of 5 μ M AA at the modified electrode retained 97.1% of its initial activity.

The reproducibility of the electrode's performance was determined. Three Co_3O_4 -GPE electrodes were prepared independently following the same method. The measurements of their peak oxidation currents at 5 μ M AA showed consistent results, with a low RSD of 4.93%. This demonstrates that the Co_3O_4 -GPE electrode has good reproducibility.

4. REAL SAMPLE ANALYSIS

The Co_3O_4 -GPE electrode was used to analyze AA in Vitamin C tablets (Pharimexco Viet Nam) via the standard addition method to assess its applicability. Table 2 summarizes the results, confirming the electrode's effectiveness for AA determination in pharmaceuticals. The measured mean AA concentration demonstrated agreement with the labeled value, and recovery rates ranged from 97.82% to 99.5%.

Table 2. Results from the analysis of AA in a real Vitamin C tablet sample.

Sample	Added (μM)	Found (μM)	Recovery (%)	RSD (%)
Vitamin C (500 mg)	3	2.96 ± 0.02	98.66 ± 0.84	0.85

5. CONCLUSION

The synthesis of Co₃O₄ porous crystalline material was performed using the ZIF-67 material as a precursor. The resulting Co₃O₄ possesses an internal hollow structure and a surface exhibiting porosity. The modified electrode developed with Co₃O₄ porous crystalline material offers high sensitivity and a low detection limit, making it promising for AA detection. It has also been successfully used to determine AA in real samples.

Acknowledgements

This research is conducted within the framework of Science and Technology Projects at institutional level of Quy Nhon University under the project code T2024.832.03.

REFERENCES

1. M. Levine, Y. Wang, S. J. Padayatty, J. Morrow, A new recommended dietary allowance of vitamin C for healthy young women, *Proceeding of the National Academy of Science*, **2001**, 98, 9842–9846.
2. C. M. Nam, K. W. Oh, K. H. Lee, S. H. Jee, S. Y. Cho, W. H. Shim, I. Suh, Vitamin C intake and risk of ischemic heart disease in a population with a high prevalence of smoking, *Journal of the American College of Nutrition*, **2003**, 22, 372–378.
3. A. Jain, A. Chaurasia, K. K. Verma. Determination of ascorbic acid in soft drinks, preserved fruit juices and pharmaceuticals by flow injection spectrophotometry: matrix absorbance correction by treatment with sodium hydroxide, *Talanta*, **1995**, 42, 779–787.
4. M. Tabata, H. Morita. Spectrophotometric

determination of a nanomolar amount of ascorbic acid using its catalytic effect on copper(II) porphyrin formation, *Talanta*, **1997**, 44, 151-157.

5. J. Lykkesfeldt. Determination of ascorbic acid and dehydroascorbic acid in biological samples by high-performance liquid chromatography using subtraction methods: reliable reduction with tris [2-carboxyethyl] phosphine hydrochloride, *Analytical Biochemistry*, **2000**, 282, 89–93.
6. R. Thomis, E. Roets, J. Hoogmartens. Analysis of tablets containing aspirin, acetaminophen, and ascorbic acid by high-performance liquid chromatography, *Journal of Pharmaceutical Sciences*, **1984**, 73, 1830–1833.
7. P. O. Barrales, M. L. F. d. Córdova, A. M. Díaz. Indirect determination of ascorbic acid by solid-phase spectrophotometry, *Analytica Chimica Acta*, **1998**, 360, 143-152.
8. M. Tabata, H. Morita. Spectrophotometric determination of a nanomolar amount of ascorbic acid using its catalytic effect on copper(II) porphyrin formation, *Talanta*, **1997**, 44, 151–157.
9. W. S. Law, P. Kubáň, J. H. Zhao, S. F. Y. Li, P. C. Hauser. Determination of vitamin C and preservatives in beverages by conventional capillary electrophoresis and microchip electrophoresis with capacitively coupled contactless conductivity detection, *Electrophoresis*, **2005**, 26, 4648–4655.
10. T. Wu, Y. Guan, J. Ye. Determination of flavonoids and ascorbic acid in grapefruit peel and juice by capillary electrophoresis with electrochemical detection, *Food Chemistry*, **2007**, 100, 1573–1579.
11. Y. Zheng, H. Zhang, L. Fu. Preparation gold nanoparticles using herb leaf extract for electro-oxidation determination of ascorbic acid, *Inorganic and Nano-Metal Chemistry*, **2018**, 48, 449-453.

12. A. Pardakhtya, S. Ahmadzadeha, S. Avazpoura, V. K. Gupta. Highly sensitive and efficient voltammetric determination of ascorbic acid in food and pharmaceutical samples from aqueous solutions based on nanostructure carbon paste electrode as a sensor, *Journal of Molecular Liquids*, **2016**, 216, 387-391.
13. X. Chen, J. Cheng, Q. Shou, F. Liu, X. Zhang. Effect of calcination temperature on the porous structure of cobalt oxide micro-flowers, *CrystEngComm*, **2012**, 14, 1271-1276.
14. C. Sun, S. Rajasekhara, Y. Chen, J. B. Goodenough. Facile synthesis of monodisperse porous Co_3O_4 microspheres with superior ethanol sensing properties, *Chemical Communications*, **2011**, 48, 12852-12854.
15. C. Sun, X. Su, F. Xiao, C. Niu, J. Wang. Synthesis of nearly monodisperse Co_3O_4 nanocubes via a microwave-assisted solvothermal process and their gas sensing properties, *Sensors and Actuators B: Chemical*, **2011**, 157, 681-685.
16. R. He, H. Liang, C. Li, J. Bai. Enhanced photocatalytic hydrogen production over $\text{Co}_3\text{O}_4@\text{g-C}_3\text{N}_4$ p-n junction adhering on one-dimensional carbon fiber, *Colloids and Surface A: Physicochemical and Engineering Aspects*, **2020**, 586, 124200.
17. H. Yu, J. Xu, Z. Liu. Functionalization of sheet structure MoS_2 with $\text{CeO}_2\text{-Co}_3\text{O}_4$ for efficient photocatalytic hydrogen evolution, *Journal of Materials Science*, **2018**, 53, 15271-15284.
18. Y. Y. Zheng, C. X. Li, X. T. Ding, Q. Yang, Y. M. Qi, H. M. Zhang, L. T. Q. Chin. Continuous synthesis for zirconium metal-organic frameworks with high quality and productivity via microdroplet flow reaction, *Chemistry Letters*, **2017**, 28, 1473-1478.
19. M. Z. Hussain, Z. Yang, Z. Huang, Q. Jia, Y. Zhu, Y. Xia. Recent advances in metal-organic frameworks derived nanocomposites for photocatalytic applications in energy and environment, *Advanced Science*, **2021**, 14, 2100625.
20. W. Li, X. Wu, N. Han, J. Chen, X. Qian, Y. Deng, W. Tang, Y. Chen. MOF-derived hierarchical hollow ZnO nanocages with enhanced low-concentration VOCs gas sensing performance, *Sensors and Actuators B: Chemical*, **2016**, 225, 158-166.
21. K. Tao, X. Han, Q. Yin, D. Wang, L. Han, L. Chen. Metal-organic frameworks derived porous In_2O_3 hollow nanorod for high-performance ethanol gas sensor, *ChemistrySelect*, **2017**, 33, 10918-10925.
22. Y. Cai, G. Fang, J. Zhou, S. Liu, Z. Luo, A. Pan, G. Cao, S. Liang. Metal-organic framework-derived porous shuttle-like vanadium oxides for sodium-ion battery application, *Nano Research*, **2018**, 11, 449-463.
23. M. H. Yap, K. L. Fow, G. Z. Chen. Synthesis and applications of MOF-derived porous nanostructures, *Green Energy & Environment*, **2017**, 3, 218-245.
24. C. Zhang, W. Chu, R. Jiang, L. Li, Q. Yang, Y. Cao, J. Yan. ZIF-67, derived hollow structured Co_3O_4 nanocatalysts: tunable synthetic strategy induced enhanced catalytic performance, *Catalysis Letters*, **2019**, 149, 3058-3065.
25. X. Yan, L. Tian, M. He, X. Chen. Three-dimensional crystalline/amorphous $\text{Co/Co}_3\text{O}_4$ core/shell nanosheets as efficient electrocatalysts for the hydrogen evolution reaction, *Nano Letters Journal*, **2015**, 15, 6015-6021.
26. T. T. H. Ngo, T. H. Y. Pham, N. T. Dang, T. V. H. Nguyen, T. H. Pham. Graziella liana turdean, graphite paste electrode modified with zeolitic imidazolate framework (ZIF-67) for the determination of acetaminophen, *Studia UBB Chemia*, **2023**, 1, 7-17.
27. K. Dhara, R. M. Debiprosad. Review on nanomaterials-enabled electrochemical

- sensors for ascorbic acid detection, *Analytical Biochemistry*, **2019**, 586, 113415.
28. A. M. Pisoschi, A. Pop, A. I. Serban, C. Fafaneata. Electrochemical methods for ascorbic acid determination, *Electrochimica Acta*, **2014**, 121, 443-460.
 29. N. Liu, M. Tang, C. Jing, W. Huang, P. Tao, X. Zhang, J. Lei, L. Tang. Synthesis of highly efficient Co_3O_4 catalysts by heat treatment ZIF-67 for CO oxidation, *Journal of Sol-gel Science and Technology*, **2018**, 88, 163-171.
 30. Y. Lu, W. Zhan, Y. He, Y. Wang, X. Kong, Q. Kuang, Z. Xie, L. Zheng. MOF-templated synthesis of porous Co_3O_4 concave nanocubes with high specific surface area and their gas sensing properties, *ACS Applied Materials & Interfaces*, **2014**, 6, 4186-4195.
 31. L. Zhang, J. Feng, K. C. Chou, L. Su, X. Hou. Simultaneously electrochemical detection of uric acid and ascorbic acid using glassy carbon electrode modified with chrysanthemum-like titanium nitride, *Journal of Electroanalytical Chemistry*, **2017**, 803, 11-18.
 32. X. Zhang, S. Yu, W. He, H. Uyama, Q. Xie, L. Zhang, F. Yang. Electrochemical sensor based on carbon-supported NiCoO_2 nanoparticles for selective detection of ascorbic acid, *Biosensors and Bioelectronics*, **2014**, 55, 446-451.
 33. F. A. Harraz, M. Faisal, A. E. A. Salami, A. M. E. Toni, A. A. Almadiy, S. A. A. Sayari, M. S. A. Assiri. Silver nanoparticles decorated stain-etched mesoporous silicon for sensitive, selective detection of ascorbic acid, *Materials Letters*, **2019**, 234, 96-100.
 34. A. Ejaz, S. Jeon. A highly stable and sensitive GO-XDA- Mn_2O_3 electrochemical sensor for simultaneous electrooxidation of paracetamol and ascorbic acid, *Electrochimica Acta*, **2017**, 245, 742-751.
 35. C. Wang, J. Du, H. Wang, C. E. Zou, F. Jiang, P. Yang, Y. Du. A facile electro chemical sensor based on reduced graphene oxide and Au nanoplates modified glassy carbon electrode for simultaneous detection of ascorbic acid, dopamine and uric acid, *Sensors and Actuators B: Chemical*, **2014**, 204, 302-309.



© 2025 by the authors. This Open Access Article is licensed under the Creative Commons Attribution-NonCommercial 4.0 International (CC BY-NC 4.0) license (<https://creativecommons.org/licenses/by-nc/4.0/>).

Chế tạo hạt nano alginate-chitosan chứa ovalbumin bằng phương pháp phun điện đồng trục để định hướng phân phối protein

Cao Lưu Ngọc Hạnh^{1,2,*}, Nguyễn Tường Vy², Phạm Duy Toàn³,
Nguyễn Thị Bích Thuyền¹, Đặng Huỳnh Giao¹, Lê Trường Thịnh²

¹Khoa kỹ thuật Hóa học, Trường Đại học Cần Thơ, Việt Nam

²Phòng thí nghiệm vật liệu composite, Khoa kỹ thuật Hóa học, Trường Đại học Cần Thơ, Việt Nam

³Bộ môn Khoa học sức khỏe, Khoa Khoa học Tự nhiên, Trường Đại học Cần Thơ, Việt Nam

Ngày nhận bài: 10/03/2025; Ngày sửa bài: 18/06/2025;

Ngày nhận đăng: 01/08/2025; Ngày xuất bản: 28/10/2025

TÓM TẮT

Liệu pháp protein đã phát triển vượt trội như liệu pháp hiệu quả và độc đáo vì những lợi ích to lớn của chúng trong lĩnh vực dược phẩm. Nghiên cứu này đã phát triển các hạt nano alginate-chitosan (ALG-CHI) chứa ovalbumin (OVA) bằng cách sử dụng điện phun đồng trục mà không cần tác nhân liên kết chéo để cung cấp protein OVA qua đường uống để định hướng liệu pháp protein. Các thông số tối ưu bao gồm điện áp 21 kV, lưu lượng 0,1/0,2 mL.h⁻¹ (lõi/vỏ), khoảng cách thu mẫu 10 cm, nồng độ polyme 4 wt% CHI và 0,5 wt% ALG. Các hạt nano ALG-OVA@CHI thu được có hình cầu, bề mặt nhẵn, không kết tụ và cấu trúc lõi - vỏ được quan sát thấy bằng hình ảnh SEM và TEM. Các hạt nano có đường kính thủy động là $313,0 \pm 129,9$ nm và thế zeta là +1,6 mV ở pH trung tính, được đo bằng thiết bị DLS. Đáng chú ý là các hạt nano ALG-OVA@CHI đã bao bọc OVA ở mức cao với hiệu suất $99,04 \pm 0,14\%$, bảo vệ OVA khỏi axit dạ dày và tạo điều kiện cho OVA giải phóng kéo dài trong điều kiện đại tràng mô phỏng trong ít nhất 8 ngày. Việc giải phóng các hạt nano này chủ yếu được kiểm soát bởi sự khuếch tán của OVA và sự phân hủy polyme trong môi trường giải phóng, theo cơ chế vận chuyển không phải Fickian của Korsmeyer-Peppas. Những phát hiện này cho thấy thiết kế ALG-OVA@CHI có tiềm năng trở thành một công thức uống để cung cấp protein.

Từ khóa: Alginate, chitosan, phun điện đồng trục, hạt nano, ovalbumin.

*Tác giả liên hệ chính.

Email: clnhanh@ctu.edu.vn

Fabrication of ovalbumin-loaded alginate-chitosan nanoparticle using coaxial electrospray for protein delivery orientation

Cao Luu Ngoc Hanh^{1,2,*}, Nguyen Tuong Vy², Pham Duy Toan³,
Nguyen Thi Bich Thuyen¹, Dang Huynh Giao¹, Le Truong Thinh²

¹Faculty of Chemical Engineering, Can Tho University, Vietnam

²Composite Materials Lab, Faculty of Chemical Engineering, Can Tho University, Vietnam

³Department of Health Sciences, College of Natural Sciences, Can Tho University, Vietnam

Received: 10/03/2025; Revised: 18/06/2025;

Accepted: 01/08/2025; Published: 28/10/2025

ABSTRACT

Protein therapeutics have emerged as effective and unique therapies because of their vast benefits in the pharmaceutical field. This study developed ovalbumin (OVA) loaded alginate-chitosan (ALG-CHI) nanoparticles using coaxial electrospray without a cross-linking agent to orally deliver OVA protein for protein therapeutics orientation. The optimal parameters include a voltage of 21 kV, a flow-rate of 0.1/0.2 mL.h⁻¹ (core/shell), a working distance of 10 cm, polymer concentrations of 4 wt% CHI and 0.5 wt% ALG. The obtained ALG-OVA@CHI nanoparticles had a spherical shape, smooth surface, no aggregation, and core-shell structure observed by SEM and TEM images. The nanoparticles had a hydrodynamic diameter of 313.0 ± 129.9 nm and a zeta potential of +1.6 mV at neutral pH, as measured by DLS equipment. Remarkably, the ALG-OVA@CHI nanoparticles highly encapsulated OVA with $99.04 \pm 0.14\%$ efficiency, protected it from gastric acid in the stomach, and facilitated sustained release OVA in the simulated colonic condition for at least 8 days. The release of these nanoparticles were primarily controlled by the diffusion of OVA and polymer disintegration in the release media, followed the Korsmeyer-Peppas non-Fickian transport mechanism. These findings suggested that the ALG-OVA@CHI design had potentials as an oral formulation for protein delivery.

Keywords: *Alginate, chitosan, coaxial electrospray, nanoparticle, ovalbumin.*

1. INTRODUCTION

Protein therapeutics have been rapidly developed in the pharmaceutical industry due to their efficacy in both diseases preventions and treatments by modulating the physiological or pathological processes of their targets.¹⁻² Proteins' primary pharmacological activities in therapeutic processes include i) replacing a deficient or abnormal protein, ii) augmenting

an existing pathway, iii) providing a novel function or activity, iv) interfering with a molecule or organism, and v) delivering a payload such as a radionuclide, cytotoxic drug, or protein effector.³ Many potentially fatal diseases, including cancer, cardiovascular disease, and diabetes, have been effectively treated by proteins.² Moreover, proteins possess high specificity, selectivity, potency, and low toxicity, which are less likely to interfere with

*Corresponding author.

Email: clnhanh@ctu.edu.vn

normal biological processes and cause adverse effects.⁴⁻⁵ Furthermore, macromolecular proteins have a significantly larger surface area for molecular binding, allowing access to a much broader range of protein targets while reducing the possibility of off-target effects.⁶ Protein therapeutics are most commonly administered via parenteral administrations such as injection or intravenous infusion.^{1,7} The routes, however, contain several drawbacks including being expensive, painful, inconvenient, and requiring professional healthcare staff. Therefore, there has recently been a shift toward producing needle-free biopharmaceuticals such as the oral protein therapeutics.⁷

For this reason, developing effective oral formulations and increasing intact protein absorption into the intestinal mucosa is currently a major focus.⁸ Furthermore, the oral route allows protein treating expression at specific locations, particularly mucosal sites, resulting in accurate and long-lasting therapy. However, direct oral protein delivery faces a number of challenges, including protein degradation in the extreme environment of the gastrointestinal tract, uncontrolled protein release, and ineffective mucosal permeability, all of which contribute to comparatively poor performance when compared to injectable administration.⁹ To enhance the treatment efficacy, the proteins must be able to pass through several physicochemical and biological barriers in the digestive tract, while also providing adequate drug concentrations at the disease state. To address these challenges, biopolymer-based nanoparticles have been used as protein delivery carriers to stabilize proteins against external obstacles and improve the bioavailability of entrapped proteins in the gastrointestinal tract.^{4,10}

Alginate (ALG) is a natural polyanionic (due to carboxylic groups) biopolymer that has been prioritized as a protein carrier due to its interesting biopharmaceutical properties, such as pH sensitivity, biocompatibility, biodegradability, muco-adhesiveness, no

toxicity, and no immunogenicity.¹¹ ALG has been extensively investigated and used in a variety of biomedical delivery systems.¹²⁻¹⁴ However, ALG has a gel structure that is typically sensitive to chelating compounds such as phosphate, citrate, ethylenediaminetetracetic acid (EDTA), and lactate, as well as anti-gelling cations found in the gastrointestinal tract.¹⁵ As a result, it is necessary to improve its stability by coating the ALG nanoparticles with another positively charged polymer such as chitosan (CHI).

CHI is a biopolymer with beneficial biological properties such as biocompatibility with protein antigens, good biodegradability, and low toxicity.¹⁶⁻¹⁸ Moreover, CHI has excellent mucoadhesive properties due to electrostatic interactions between positively charged polymers and negatively charged mucosal surfaces, as well as advanced penetration ability across the colonic wall.¹⁹ CHI is one of the most promising positively charged coating materials for improving the stability of encased proteins. As a result, coating ALG nanoparticles with CHI (ALG@CHI) could form a polyelectrolyte complex through ionic interaction, which was expected to reduce porosity, encapsulated protein leakage, and increase stability across wide pH ranges and enzymatic activities.¹⁵ Ling et al. developed ALG-CHI multilayer nanoparticles, which delivered protein AvrA, a protein with dual anti-inflammatory and anti-apoptotic enzymatic function, for colitis therapy. The results indicated that the nanoparticles delivered protein to intestinal epithelia and reduced inflammation in a murine colitis model.²⁰ Chen et al. demonstrated effective insulin encapsulation of pH-sensitive ALG-CHI nanoparticles for prolong insulin release to diabetic rats orally and reducing blood glucose levels effectively.²¹

In this study, efforts were made to develop an oral delivery system that loaded ovalbumin (OVA), a model protein, with CHI and ALG, resulting in ALG-OVA@CHI nanoparticles. OVA has been widely utilized in protein therapeutics research due to its convenient stability at room

temperature and cost effectiveness.²¹ From the viewpoint of protein structure, OVA contain disulfide links in their molecular chains; as is known, the disulfide linkages have a remarkable antioxidation-reduction function.²² The OVA-loaded polymeric nanoparticles are fabricated through the advanced coaxial electrospraying (CES) method. The CES technique is a simple one-step method for encapsulating proteins, which involves applying a high electric field to the capillary needle. The proteins and organic solvents flow through the internal capillary needle, while polymer solutions flow through the external capillary needle. The high electric charges in the liquid droplet at the needle tip result in the formation of nanoparticles via Coulomb fission.²³ The advantage of this method is that it is possible to prepare nanoparticles without the use of cross-linking agents. Therefore, the produced nanoparticles can robustly release the OVA protein in an appropriate environment, supplying the required dosage, allowing the therapy to take place quickly within a specific time. Furthermore, the biodegradable polymeric system can be safely removed from the body without causing harm in the long term, whereas the cross-linker might remain in the body for an extended period. Besides, compared to other nanoparticle production methods, the CES method is widely used for protein nanoparticle preparation because it reduces protein denaturation by using less solvent. This method's other advantages include ease of processing, high drug encapsulation, and a narrower range of size distribution.^{25,26} As a result, CES technology is expected to produce uniform ALG-OVA@CHI nanoparticles with high encapsulation efficiency and controllable release properties in the gastrointestinal tract.

2. MATERIALS AND METHODS

2.1. Materials

OVA (78%), phosphate buffer saline (PBS, pH 7.4) were offered by Himedia, India. CHI (deacetylation of 92%) was supplied by Cemaco company, Vietnam. ALG sodium salt (low

viscosity, ≤ 200 mPa.s) was purchased from Shanghai Zhanyun Chemical, China. Acetic acid (CH_3COOH , 99.5%), sodium hydroxide (NaOH, 99%), and hydrochloric acid (HCl, 37%) were obtained from Xilong, China. All other chemicals were used without further purification.

2.2. Preparation of the material solutions

The core solutions were prepared by dissolving accurate amounts of ALG (0.05 g, 0.1 g, and 0.15 g) in 10 mL of distilled water under stirring at 60 rpm for 6 h at room temperature to get 0.5 wt%, 1 wt%, and 1.5 wt% ALG solutions, respectively. Subsequently, 0.0125 g OVA was added into the ALG solutions and stirring at 60 rpm for 3 h at room temperature to attain the homogeneous core solutions. The 4 wt% CHI shell solution was obtained by adding 0.4 g CHI into 10 mL of 90% (v/v) acetic acid under agitating at 60 rpm within 24 h at room temperature.

2.3. Fabrication of ALG-OVA@CHI nanoparticles

The ALG-OVA@CHI nanoparticles were fabricated by a single-step CES method, using equipment consisted of a high-voltage supply (GENVOLT 73030, USA), syringe pumps, a stainless steel capillary needle, 1-mL plastic syringes, and aluminum nanoparticle collector.

Briefly, the prepared core solutions (0.0125 g OVA + 0.5 wt%/1 wt%/1.5 wt% ALG) and shell solution (4 wt% CHI) were injected into syringes that connected to coaxial capillary needle. The high voltage (19, 21, and 23 kV) were applied to the capillary needle tip. The distance between the needle tip and the collector (10, 13, and 16 cm) were investigated. The core and shell solutions flow rate was at 0.1 mL.h^{-1} and 0.2 mL.h^{-1} , respectively.²⁷ During the CES process, the droplets moved to collector while the solvent was allowed to evaporate and the nanoparticles were acquired at the collector. Successively, the obtained nanoparticles were dispersed in absolute ethanol to remove the free OVA.²⁸ Afterward, centrifugation was conducted

at 10,000 rpm for 15 min at 5°C to collect the nanoparticles. The particles were then placed in petri dishes and left to undergo solvent evaporation at room temperature, resulting in the final nanoparticles product.

2.4. Characterizations of physicochemical properties

Scanning electron nanoscopy (SEM, Hitachi S-4800, Japan) and an optical nanoscope (Nikon EPIPHOT 200, Japan) was used to investigate the shape and surface morphology of two different types of nanoparticles. Samples were sputter coated with a conductive layer of platinum for SEM analysis, then scanned in a low vacuum chamber with a focused electron beam at 10 kV and magnifications of 10,000 and 60,000. As the image formed, secondary electrons emitted by the samples were detected. Transmission electron nanoscopy (TEM) was used to investigate the nanostructure of ALG-OVA@CHI particles on the JEM-1400 Flash in Jeol, Japan, at an acceleration voltage of 120 kV and magnification of 80,000 in a vacuum environment. To prepare the sample for measurement, the nanoparticle suspensions were diluted in ethanol and placed on a cellulose net.

The particle size and zeta potential of both nanoparticles were determined using the dynamic light scattering (DLS) technique in a Zetasizer on SZ-100Z2, Horiba, Japan. A 5 mL sample was prepared in alcohol and measured at a holder temperature of 25°C and a scattering angle of 90°. The results were expressed as mean $\pm$ standard deviation, with a zeta potential dimension of mV.

To investigate the chemical structures and interactions between components in nanoparticles, a Fourier transform infrared spectrometer (FTIR Nicolet 6700, Thermo Fisher Scientific, USA) was used to record FTIR spectra of samples. The scanning parameters were a wavelength range of 4000-400 cm^{-1} and a resolution of 4 cm^{-1} . The spectra samples show

their characteristic peaks in the wavenumber range.

2.5. Determination of OVA encapsulation efficiency

The encapsulation efficiency of the ALG-OVA@CHI nanoparticles was assessed by dispersing 10 mg of the particles in 20 mL of distilled water and executing ultrasonic process for 280 min. Every 40 min, 2 mL of the sample was centrifuged at 10,000 rpm for 3 min at 5°C^{29,30} to collect the supernatant and the nanoparticles were re-dispersed in water to continue the experiment. From the resulting supernatant, 1 mL was mixed with 2 mL of Bradford reagent and allowed to react for 3 min. The concentration of OVA was determined using UV-Vis spectroscopy method at a detection wavelength of 595 nm, which based on the standard curve equation of $y = 7.6119x + 0.7886$ ($R^2 = 0.9977$). Each determination was conducted in triplicate, the encapsulation efficiency (H%) was calculated using the formula (1), and the results were displayed in term of means $\pm$ standard deviation (SD).

$$H\% = \frac{\text{Encapsulated mass of OVA}}{\text{Total mass of OVA}} \times 100 \quad (1)$$

2.6. In vitro OVA release test

The release behavior of OVA from ALG-OVA@CHI nanoparticles was investigated in PBS solution with different pH values of 1.2 and 7.4, simulating the gastric and colonic parts of the digestive tract, respectively.^{31,32} Briefly, 10 mg of the dried nanoparticle was immersed in 20 mL release medium, which were incubated at $37 \pm 1^\circ\text{C}$ within 8 days. At the predefined time intervals, 1 mL of sample were taken out from the release solution and centrifuged, then, it was mixed with 2 mL Bradford reagent for protein quantification at 595 nm.^{29,30} To the release media, 1 mL of fresh buffer medium were returned. The amount of OVA protein released were calculated based on the calibration curve of $y = 7.5107x + 0.7508$ ($R^2 = 0.9966$) for pH 1.2 and $y = 8.1714x + 0.7458$ ($R^2 = 0.9952$) for pH 7.4. All measurements were

performed in triplicate for reliable data and the cumulative OVA release was calculated using the following formula (2):

$$\text{Cumulative release (\%)} = \frac{C_t V_0 + V \sum_{i=1}^{t-1} C_i}{M_0 - \sum_{i=1}^{t-1} M_i} \times 100 \quad (2)$$

Where C_t , C_i were the released OVA concentrations at the time point t and i , V_0 was the total volume, V was the withdrawal volume (1 mL), and M_0 , M_i were the initial OVA amount and withdrawal OVA amount at the time point i .

Release kinetic models

The general mathematical models were employed to evaluate the protein release kinetics, including the zero-order (3), first-order (4), Higuchi (5) and Korsmeyer-Peppas (6) kinetic models.

Zero-order kinetic model: indicates that the rate of protein release is independent of the protein concentration in the nanoparticles. In another words, constant amount of protein is released in a time-dependent manner. Model expression:

$$M_t = M_0 + k_0 t \quad (3)$$

First-order kinetic model: describes a release rate proportional to the amount of drug remaining in the nanoparticles, or the release rate decreases as the protein concentration decreases over time. Model expression:

$$\ln M_t = \ln M_0 - k_1 t \quad (4)$$

Higuchi kinetic model: argues that the rate of drug release is related directly to the concentration gradient within the nanoparticles by expressing the drug release amount as a square root of a time-dependent process. Model expression:

$$M_t = k_H t^{\frac{1}{2}} \quad (5)$$

Korsmeyer-Peppas kinetic model:

characterizes the mechanism of protein release from the polymeric nanoparticle systems, including diffusion, swelling, erosion or a complex release profile of an initial burst release followed by a sustained release. Model expression:

$$\frac{M_t}{M_0} = k_{KP} t^n \quad (6)$$

Where M_0 , M_t are the initial amount of OVA and the released OVA at the time point t ; whereas k_0 , k_1 , k_H , and k_{KP} are the release constants of the zero-order, first-order, Higuchi and Korsmeyer-Peppas models, respectively. In Korsmeyer-Peppas model, the n value is the release exponent indicative of the mechanism of transport of the protein through the polymeric matrix, in which $0 < n < 0.45$, it is Fickian diffusion, $0.45 < n < 0.89$ is non-Fickian diffusion or abnormal release, and $n > 0.9$ is skeleton dissolution mechanism.

3. RESULTS AND DISCUSSIONS

3.1. Factors affecting ALG-OVA@CHI nanoparticles formation

To find the optimal formulating condition, this study aimed to comprehend how applied voltage, ALG concentration, and injector-to-collector distance affect the morphology and size of the ALG-OVA@CHI nanoparticles. The examined parameters included voltage levels (19, 21, and 23 kV), ALG concentrations (0.5 wt%, 0.1 wt%, and 1.5 wt% w/v), and working distances (10, 13, and 16 cm).

3.1.1. Effect of the applied voltage

The effect of different voltage levels (19, 21, and 23 kV) on the SEM morphology of ALG-OVA@CS nanoparticles were firstly investigated (Figure 1). To that end, the Taylor cone plays a crucial role in the CES process as it enables the formation of uniform and stable droplets, which ultimately determine the morphology and size of the nanoparticles.

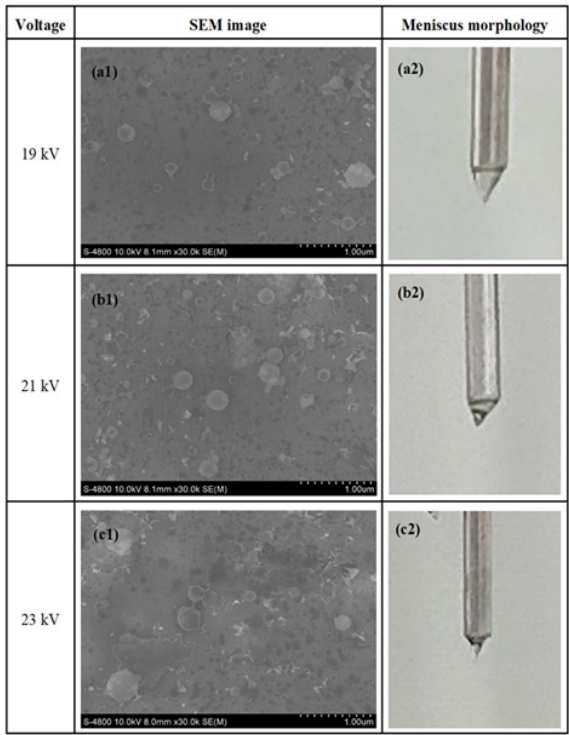


Figure 1. SEM images of ALG-OVA@CHI nanoparticles and meniscus morphology at different voltage of 19, 21, and 23 kV.

At 19 kV, the prepared nanoparticles had a nearly spherical shape but possessed an uneven size distribution (Figure 1a). This was ascribed to the low imposed voltage that leads to insufficient electrical force to overcome the hydrodynamic forces, including the viscous force, the surface tension, and the frictional force.³³ The imbalance of these forces caused the formation of unstable conical meniscus at the needle tip.³⁴ In this case, the CES mode was considered as the nano-dripping mode, resulting in heterogeneous nanoparticles.³⁵ Increasing the voltage to 21 kV produced spherical nanoparticles and smooth surfaces with relative more uniform size (Figure 1b). It was attributed to the more balanced forces that formed a stable Taylor cone. In addition, the interface between the inner and outer solutions was obvious during this status, contributing to the establishment of the stable cone-jet mode. However, when the voltage was further increased to 23 kV, the nanoparticles had rough surfaces and did not achieve a fully spherical

shape (Figure 1c). It was explained that the surface tension was substantially reduced while the electric field dominated the hydrodynamic force, resulting in the stretching of meniscus tip. Furthermore, the interface was vibratile and difficult to observe, suggesting that the pulsed cone-jet regime was created at 23 kV. As a result, the findings indicated that a voltage of 21 kV was ideal for obtaining ALG-OVA@CHI nanoparticles with desired properties.

3.1.2. Effect of the ALG concentration

ALG concentration plays an important role in the protection and sustained release of proteins by polyelectrolyte complexation with CHI. The nanoparticles produced at an ALG concentration of 0.5 wt% had a spherical shape along with secondary droplets appearance (Figure 2a), which proved that the Taylor cone was successfully created at this concentration. However, undesirable results attained when the ALG concentrations increased to 1 wt% and 1.5 wt% (Figure 2b, 2c), which yielded sparse-and-undeveloped nanoparticles.

Table 1. Mean diameter and PDI value of ALG-OVA@CHI nanoparticles at different ALG concentrations with the fixed voltage of 21 kV and solution flow rate of 0.1/0.2 mL/h (core/shell).

ALG concentration (wt%)	Mean diameter (nm)	PDI
0.5	313.0 ± 129.9	0.202
1	393.5 ± 172.7	0.395
1.5	398.3 ± 195.1	0.022

At higher ALG concentrations, the solution become more viscous, and the higher viscosity leads to the requirement of more energy to spray the polymer solution from the meniscus.³⁶ Therefore, while the imposed energy was kept constant, it was inefficiently to transform the liquid into nanoparticles. Additionally, as the

ALG concentrations increased, the gravity of polymer solution boosted up abundantly, thereby the meniscus tended to be elongated, as in the case of ALG 1.5 wt%. Moreover, the nanoparticle sizes were also increased from ~300 nm to ~400 nm, with increasing ALG concentrations (Table 1). Furthermore, all PDI values exhibit regular particle size distributions ($PDI < 0.3$). Conclusively, to produce particles of a minimum size range with narrow size distribution, the ALG concentration of 0.5 wt% was selected for future experiments.

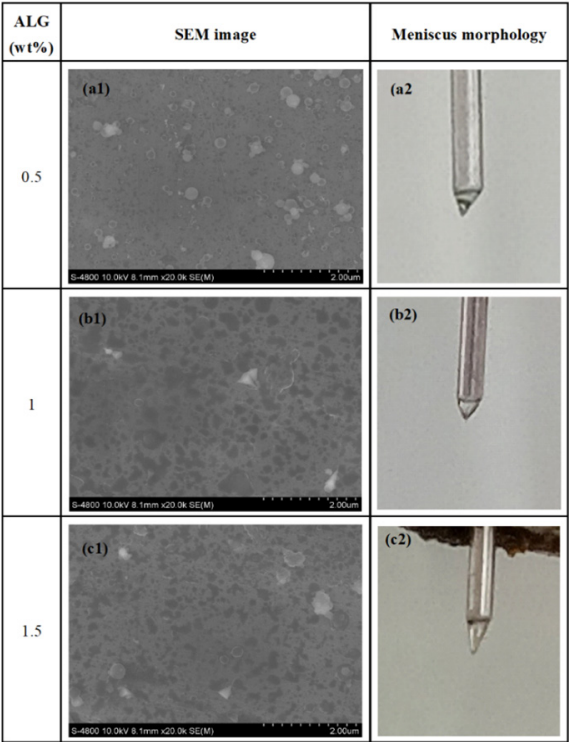


Figure 2. SEM images of ALG-OVA@CHI nanoparticles and meniscus morphology at different ALG concentration of 0.5%, 1%, and 1.5%.

3.1.3. Effect of the needle tip-to-collector distance

Besides the applied voltage and the material concentration, the needle tip-to-collector distance is another factor that plays a significant role in the performance and stability of the granulation process.^{31,37} Figure 3 demonstrates nanoparticles produced at various distance of 10, 13, and 16 cm.

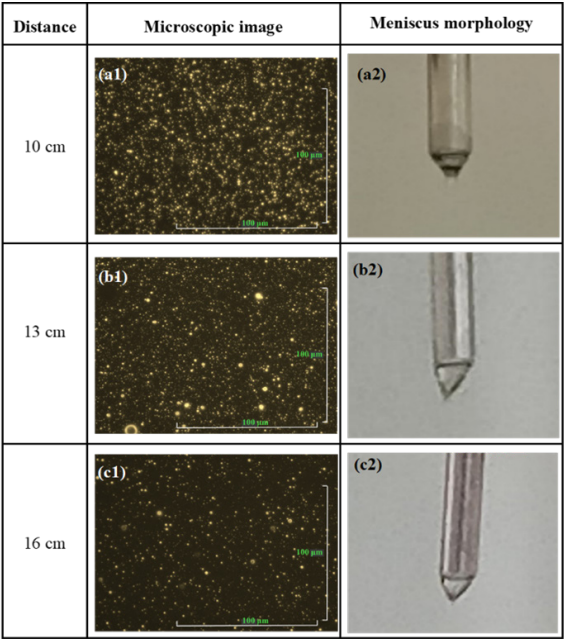


Figure 3. Nanoscope images of ALG-OVA@CHI nanoparticles and meniscus morphology at different needle tip-to-collector distances of 10, 13, and 16 cm.

When the distance was set at 10 cm, the nanoparticles acquired spherical morphology and non-aggregation property, which confirmed that this distance was adequate to accomplish complete evaporation of the solvent before reaching the collector (Figure 3a). At this distance, the Taylor cone could be kept steady, forming a steady jet generated at the tip of the cone. The distance increment to 13 cm and 16 cm has two significant effects on the CES process. Firstly, the expansion in distance caused a reduction in collected nanoparticle, as they dispersed into the surrounding environment instead of depositing in the grounded collector plate to form nanoparticles, resulting in a low electrospraying performance (Figure 3b, 3c). Secondly, the conical meniscus had an unconventional morphology that was attributed to the decreased electric field strength due to a distance increase between the electrodes (the electrical field strength is defined as the applied voltage divided by the distance between the tip and collector).³⁴ Therefore, it was convinced that increasing the distance between the injector and the collector had a direct impact on the nanoparticle collection efficiency and the

stability of the Taylor cone. In summary, the optimal needle tip-to-collector distance was set at 10 cm.

3.2. Diameter, zeta potential and structure of ALG-OVA@CHI nanoparticles

Utilizing the optimal parameter of the applied voltage of 21 kV, the flow rate of 0.1/0.2 mL.h⁻¹ (core/shell), the needle tip-to-collector distance of 10 cm, and the polymer concentrations of 4 wt% CHI and 0.5 wt% ALG, the final ALG-OVA@CHI nanoparticles were further investigated their physicochemical properties.

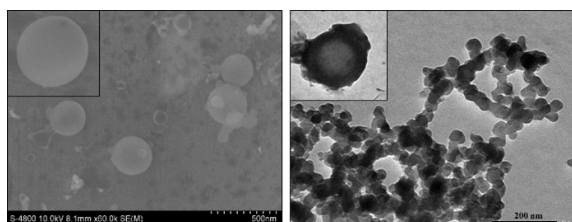


Figure 4. SEM and TEM images of ALG-OVA@CHI nanoparticles.

The interaction of orally administered nanoparticles with the mucous epithelium in the intestinal environment is critical for protein delivery. To ensure mucosal adhesion and penetration through mucous membranes of ALG-OVA@CHI nanoparticles, the diameter and zeta potential of nanoparticles synthesized under optimal conditions were determined using the DLS technique. For this, the synthesized nanoparticles had a hydrodynamic diameter of 313.0 ± 129.91 nm, which was appropriate diameter for mucus layer permeation (less than 500 nm).³⁸ Moreover, the particles possessed a zeta potential of +1.6 mV at pH 7.0, possibly due to the presence of a polycation CHI layer coating on the surface of the nanoparticles. This positively charged property allows for electrostatic interactions with the negatively charged mucus on the mucosal wall, which helps nanoparticles adhering to the mucosal surface and avoiding mucosal clearance.^{39,40} Additionally, the polydispersity index (PDI) of the nanoparticles was 0.202, indicating good monodispersity in size and stable in aqueous circumstances, which are suitable for biological applications. Furthermore,

SEM image showed nanoparticles possessed spherical shape with smooth surface, whereas TEM results showed were generally agglomerated due to the lack of dispersion medium and absence of surface steric stabilizers, however, a complete core-shell structure was observed. In the light core, ALG could interact with CHI through electrostatic interactions to form polyelectrolyte complex or hydrogel structure, and soluble OVA distributed in spaces of that polymer matrix, thus, when observed through TEM they seem appear to have a porous structure. Conversely, only CHI is presented in the dark shell in a relatively high concentration, resulting in a substantial entanglement of the polymer chains and a dense shell creation (Figure 4). This demonstrated the successful fabrication of a core-shell ALG-OVA@CHI nanoparticle structure with the single-step CES technique.

3.3. Chemical characteristics

The FTIR analysis revealed important insights about the chemical characteristics and the interactions between the components in the ALG-OVA@CHI nanoparticles (Figure 5). The FTIR spectra of CHI displayed feature peaks at 3453 cm⁻¹, 1624 cm⁻¹, 1416 cm⁻¹, 1339 cm⁻¹, and 1028 cm⁻¹, corresponding to the overlapping areas of the O–H and N–H stretching vibrations, C–H stretching, N–H bending (amide I, II), C–N stretching (amide III) and C–O–C bond, respectively.⁴¹ In the ALG spectrum, characteristic peaks at 3479 cm⁻¹, 2840 cm⁻¹, 1633 cm⁻¹, 1358 cm⁻¹, and 1045 cm⁻¹ represented the –OH group stretching vibrations, C–H stretching, anti-symmetric, symmetric stretching of carbonyl groups, and C–O–C linkage, respectively.⁴² Notably, in the ALG@CHI nanoparticles, the amide group of the cationic polymer (ALG) interacts with the carbonyl group of anionic polymer (ALG), resulting in the polyelectrolyte complex.⁴³ As a result of the interactions, the displacement of these groups were observed with a shift from 1624 cm⁻¹ to 1627 cm⁻¹ (amide groups) and from 1358 cm⁻¹ to 1397 cm⁻¹ (carbonyl groups). Additionally, the absorption at 1045–1084 cm⁻¹, relating to the elongation of C–O–C bonds, confirmed the effective interactions between CHI and ALG in their structure.

In the ALG-OVA@CHI nanoparticle spectrum, a broader and intense band centered at 3468 cm^{-1} corresponded to stretching vibrations from the overlapping areas of the O–H bond in the polymer structures and N–H of amide group in the protein structures. The peaks observed at 2921 and 2855 cm^{-1} were due to symmetric and asymmetric C–H stretching vibrations of pyranose ring.⁴⁴ Characteristic bands of CHI, ALG, and OVA were usually seen in the range 1600 – 1650 cm^{-1} , correlated with N–H bending (amide I), C–O stretching (carbonyl group) and C–O stretching (amide I), respectively. Those bands shifted and overlapped each other, creating a strong peak at 1609 cm^{-1} . The disappearance of the symmetric carbonyl group (signal at 1397 cm^{-1}), in comparison to ALG@CHI spectrum, confirmed the completely encapsulation of ALG within CHI nanoparticle. The elongation of C–O–C linkage was shifted in the ALG-OVA@CHI nanoparticle, from 1084 cm^{-1} to 1095 cm^{-1} , which reveals the successfully encapsulation of OVA protein in the nanoparticle. This result is also consistent with previous study by Cao et al.⁴⁵

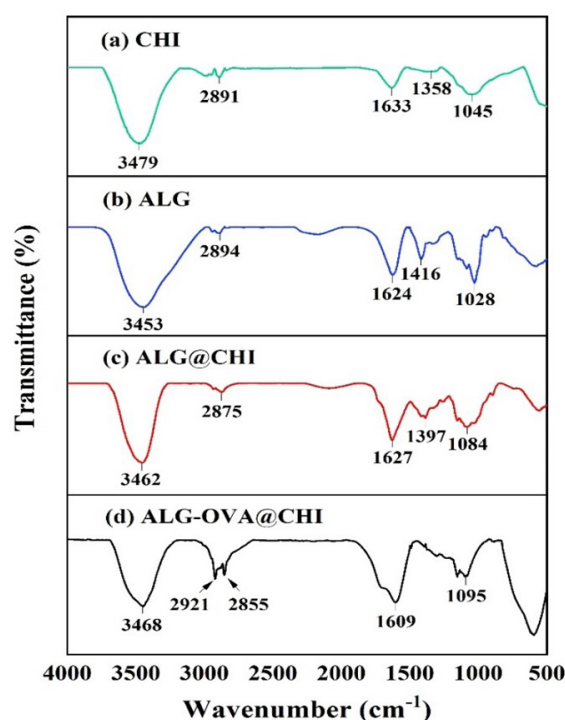


Figure 5. FTIR spectra of (a) CHI, (b) ALG, (c) ALG@CHI nanoparticle and (d) ALG-OVA@CHI nanoparticle.

3.4. Encapsulation efficiency

The encapsulation efficiency of ALG-OVA@CHI was determined using UV-Vis measurement as it directly measures the OVA protein entrapment. In this experiment, distilled water was used as medium to disperse the nanoparticles, which could dissolve OVA completely and had no interference between protein and Bradford reagent. The OVA encapsulation efficiency of the nanoparticles was $99.04 \pm 0.14\%$ (Figure 6), which was significantly higher than those in other reports.^{32,46} This result indicated that the CES technique is an effective method to fabricate core-shell nanoparticles with outstanding encapsulation efficiency. The reasons for this is that the nanoparticles preparation process happened in the air phase, not the liquid phase, thus limiting the diffusion and loss of OVA. In addition, as previously discussed, the CES technique could provide highly dispersed and distributed OVA within CHI and ALG polymer matrix, leading to a high encapsulation efficiency. Interestingly, the formation of polyelectrolyte complex between CHI and ALG in the shell of the nanoparticles were considered as a self-assembly cross-linking process, contributing to the prevention of OVA leakage. The similar results were observed in the previous studies of Manosree et al. and Zaeim et al.^{47,48}

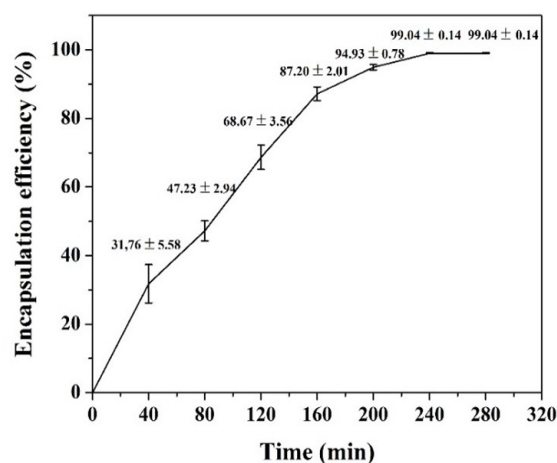


Figure 6. OVA encapsulation efficiency of ALG-OVA@CHI nanoparticles.

3.5. In vitro OVA release results

In this study, OVA protein was encapsulated within ALG-OVA@CHI nanoparticles, which were expected to both protect the drug from degradation in an acidic gastric environment (pH 1.2) and exhibit sustained OVA release in a colonic environment (pH 7.4). Furthermore, the ALG-OVA@CHI design aims to improve the nanoparticles' colon-targeting characteristics by using of polycationic CHI coating. Therefore, the in vitro protein cumulative release behaviors of ALG-OVA@CHI nanoparticles were investigated in physiological medium PBS (pH 1.2 and pH 7.4) at 37°C for 8 days (Figure 7).

During the first initial 2 h (i.e., the expected retention time of the nanoparticles in the gastric environment), as expected, the nanoparticles effectively retained OVA release in both pH 1.2 and pH 7.4 conditions by limiting protein delivery to less than 30% (Figure 7a). The complexation of CHI and ALG at pH 1.2 reduced the burst release behavior of the nanoparticles. This reduction was achieved by a decrease in vacant spaces in the polymer network, which reduced the swelling of the nanoparticles, hence limiting protein burst release.⁴⁹ In detail, the protein highly released within the first 0.5 h (25.53% for pH 1.2 and 12.02% for pH 7.4), followed by a stable release until 2 h, reaching 27.51% for pH 1.2 and 19.37% for pH 7.4. This was attributed to the free OVA's poor attachment to the nanoparticles' outermost layer/surface, resulting in rapid release. On the other hand, the encapsulated protein was released over an extended period of time, resulting in a steady release pattern for OVA. At pH 7.4, the CHI outer shell was insoluble, resulting in a dense structure. As a result, the diffusion velocity of protein out of the carrier decreased, and OVA content release in pH 7.4 was lower than that in pH 1.2. Conclusively, the ALG-OVA@CHI

design could retain the drug release and avoid the drug degradation in an acidic environment, thereby meeting the study's objective.

The OVA protein release experiment was extended to 8 days to investigate the long-term release behavior and predict structural variation (Figure 7b). To this purpose, the OVA was steadily released in both pH settings, reaching maximum values of 91.41% for pH 1.2 and 93.42% for pH 7.4 after 6 days. The gradual increase in OVA release content was attributed to the structural variation of nanoparticles that had been immersed in the release medium for an extended period of time. In the acidic condition, the solubility of the CHI layer increased significantly, causing the CHI shell to dissolve faster, resulting in strong nanoparticle degradation. As a consequence, the interaction between CHI and ALG was weakened and the repulsion between molecular chains of the polymer matrix increased, thus, the ALG@CHI carrier structure could not effectively prevent OVA diffusion into the release environment.⁵⁰ Conversely, in a light alkaline environment of pH 7.4, the hydrophilicity of the carriers increased due to interactions between hydrophilic groups in polymer structure and water molecules, causing water to potentially permeate into the nanoparticles and dissolved OVA, resulting in an increase in the percentage of cumulative release. It was also worth noting that the protein content remaining after the experiments could be the result of drug interactions with carriers. In summary, the ALG-OVA@CHI design was identified as a potential material capable of inhibiting OVA release in gastric acidic conditions, exhibiting sustained protein release in the colonic environment, and increasing OVA bioavailability in the gastrointestinal tract, all of which contributed to the development of controlled release and orally therapeutic effectiveness.

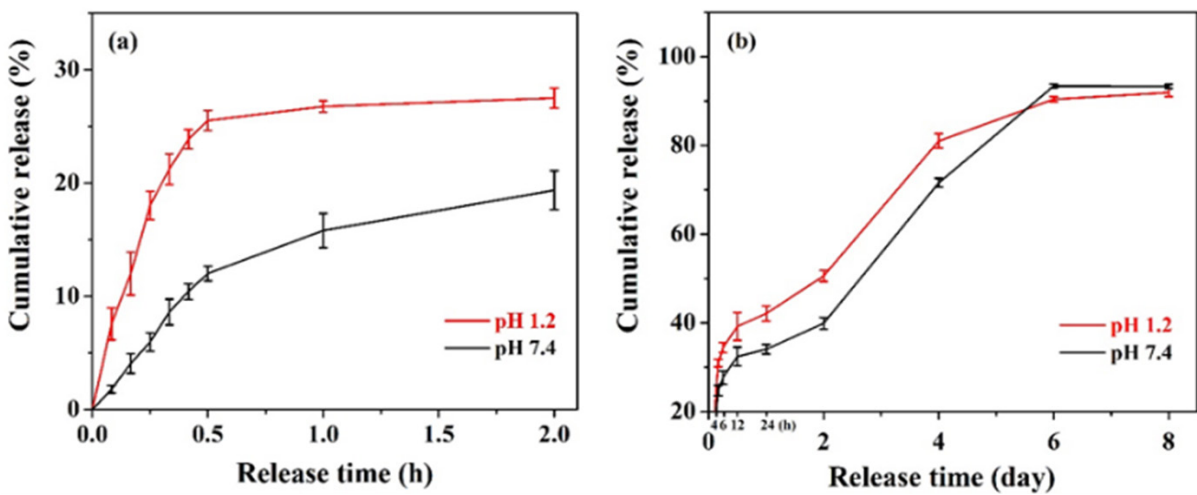


Figure 7. In vitro release profile of OVA (a) in initial 2 hours and (b) in 8 days.

Table 2. The kinetic models for the OVA release at different pH of 1.2 and 7.4 within 2 hours and 8 days.

		Zero-order		First-order		Higuchi		Korsmeyer-Peppas		
		R ²	k ₀	R ²	k ₁	R ²	k _H	R ²	k _{KP}	n
pH 1.2	2 h	0.4706	10.541	0.4969	0.1278	0.7622	20.499	0.7866	23.150	0.4078
	8 day	0.8629	0.4288	0.9683	0.0132	0.9521	5.9453	0.9772	20.482	0.7498
pH 7.4	2 h	0.8077	9.2479	0.8306	0.1040	0.9103	4.4663	0.9141	11.918	0.3962
	8 day	0.9006	0.4777	0.9512	0.0143	0.9211	10.831	0.9791	11.634	0.4774

To better understand the releasing process of OVA, the release kinetic models were studied by fitting the release data of OVA in physiological medium PBS (pH 1.2 and pH 7.4) using zero-order, first order, Higuchi and Korsmeyer-Peppas models, and the release mechanism was obtained by the kinetic parameters that characterized the models. According to the Table 2, the Korsmeyer-Peppas kinetic model had reasonably high correlation coefficient (R^2), which was considered to be consistent with the release mechanism in all stages. The release of OVA nanoparticles under simulated gastric condition exhibited higher release rates than those at simulated colonic environment (k_{KP} at pH 1.2 > k_{KP} at pH 7.4) demonstrating that the release of OVA from CHI@ALG nanoparticles was more promoted in an acidic medium, and this

assumption was compatible with the experiment as discussed in previous section. During the first phase (2 hours), the diffusion exponent (n) of the Korsmeyer-Peppas model was less than 0.45, indicating that Fickian diffusion was primarily controlling OVA release from the ALG-OVA@CHI nanoparticles. While in the entire release process (8 hours), the R^2 coefficient was better fitted with the Korsmeyer-Peppas model, and the n value was in the range of 0.45-0.89 suggesting that the OVA release followed an anomalous (non-Fickian) transport mechanism, which involved a combination of diffusion of OVA and disintegration of polymer matrix.⁵¹ Besides, when calculating first-order release kinetics, the whole release process had a high R^2 value, contributing the decrease of release rates as the OVA concentration decreased over time

corresponding to the late state of release profiles. In conclusion, the delivery system's flexibility in different phases of the release process made the ALG-OVA@CHI nanoparticles as an ideal and versatile formulation for orally protein delivery applications.

4. CONCLUSIONS

This study effectively produced an ALG-CHI nanoparticle encapsulating OVA, a model protein utilized in protein therapeutics research, utilizing the CES approach for possible protein therapies. The optimal CES parameters included an applied voltage of 21 kV, a flow rate of 0.1/0.2 mL.h⁻¹ (core/shell), a needle tip-to-collector distance of 10 cm, and polymer concentrations of 4 wt% CHI and 0.5 wt% ALG. The SEM and TEM images revealed that the ALG-OVA@CHI nanoparticles had a spherical shape, smooth surface, and a core-shell structure. The DLS measurements showed that the nanoparticles had a hydrodynamic diameter of 313.0 ± 129.9 nm. The FTIR analysis confirmed the interactions between the nanoparticle components, such as the formation of a polyelectrolyte complex between CHI and ALG, as well as the presence of OVA in the formulation. A notable feature of the ALG-OVA@CHI nanoparticle was its encapsulation efficiency OVA reached $99.04 \pm 0.14\%$. Under pH 1.2, the ALG-OVA@CHI formulation effectively shielded the encased protein from the acidic environment, while under pH 7.4, the nanoparticle demonstrated a sustained release achieving release capacity of above 90%. Additionally, the OVA protein release profiles from ALG-CHI nanoparticle primary relied in the diffusion of OVA and the dissolution of polymer network, adhering to the Korsmeyer-Peppas kinetics model. The development strategy for nanoparticles involved examining the biochemical properties of the microparticles, and further in vivo testing to assess therapeutic efficacy. In conclusion, the nanostructure of ALG-CHI could be an excellent candidate for oral OVA delivery, contributing to the advancement of protein therapeutics.

REFERENCES

1. F. Fitriagustiani, A. Z. Mustopa, S. Budiarti, M. F. Warsito, R. D. Pratiwi, D. F. Agustiyanti, M. Nurfatwa. Formulation and characterization of alginate coated chitosan nanoparticles as therapeutic protein for oral delivery system, *Trends in Sciences*, **2022**, 19(18), 5797.
2. S. J. Cao, S. Xu, H. M. Wang, Y. Ling, J. Dong, R. Xia, X. Sun. Nanoparticles: oral delivery for protein and peptide drugs, *American Association of Pharmaceutical Scientists*, **2019**, 20(190), 1-11.
3. P. J. Carter. Introduction to current and future protein therapeutics: a protein engineering perspective, *Experimental Cell Research*, **2011**, 317(9), 1261-1269.
4. R. Vaishya, V. Khurana, S. Patel, A. K. Mitra. Long-term delivery of protein therapeutics, *Expert Opinion on Drug Delivery*, **2015**, 12(3), 415-440.
5. B. Leader, Q. J. Baca, D. E. Golan. Protein therapeutics: a summary and pharmacological classification, *Nature Reviews Drug Discovery*, **2008**, 7, 21-39.
6. J. R. Kintzing, M. V. F. Interrante, J. R. Cochran. Emerging strategies for developing next-generation protein therapeutics for cancer treatment, *Trends in Pharmacological Sciences*, **2016**, 37(12), 993-1008.
7. Z. Antosova, M. Mackova, V. Kral, T. Macek. Therapeutic application of peptides and proteins: parenteral forever?, *Trends in Biotechnology*, **2009**, 27(11), 628-635.
8. J. E. V. Ramirez, L. A. Sharpe, N. A. Peppas. Current state and challenges in developing oral vaccines, *Advanced Drug Delivery Reviews*, **2017**, 114, 116-131.
9. V. T. Le, P. M. Lovanti, A. M. A. Fattah. Stabilization challenges and formulation strategies associated with oral biologic drug delivery systems, *Advanced Drug Delivery Reviews*, **2015**, 93, 95-108.

10. F. M. Almutairi. Biopolymer nanoparticles: a review of prospects for application as carrier for therapeutics and diagnostics, *International Journal of Pharmaceutical Research & Allied Sciences*, **2019**, 8(1), 25-35.
11. P. Severino, C. F. Silva, L. N. Andrade, D. L. Oliveira, J. Campos, E. B. Souto. Alginate nanoparticles for drug delivery and targeting, *Current Pharmaceutical Design*, **2019**, 25(11), 1312-1334.
12. C. Zhang, G. Shi, J. Zhang. Targeted antigen delivery to dendritic cell via functionalized alginate nanoparticles for cancer immunotherapy, *Journal of Controlled Release*, **2017**, 256, 170-181.
13. S. Mallakpour, E. Azadi, C. M. Hussain. Chitosan, alginate, hyaluronic acid, gums, and β -glucan as potent adjuvants and vaccine delivery systems for viral threats including SARS-CoV-2: A review, *International Journal of Biological Macromolecules*, **2021**, 182, 1931-1940.
14. D. Thomas, K. K. Thomas, M. S. Latha. Preparation and evaluation of alginate nanoparticles prepared by green method for drug delivery applications, *International Journal of Biological Macromolecules*, **2020**, 154, 888-895.
15. H. Liu, M. Xie, S. Nie. Recent trends and applications of polysaccharides for microencapsulation of probiotics, *Food Frontiers*, **2020**, 1(1), 45-59.
16. P. Jana, M. Shyam, S. Singh, V. Jayaprakash, A. Dev. Biodegradable polymers in drug delivery and oral vaccination, *European Polymer Journal*, **2021**, 142, 110155.
17. M. Song, L. Li, Y. Zhang, K. Chen, H. Wang, R. Gong. Carboxymethyl- β -cyclodextrin grafted chitosan nanoparticles as oral delivery carrier of protein drugs, *Reactive and Functional Polymers*, **2017**, 117, 10-15.
18. A. Sogias, A. C. Williams, V. V. Khutoryanskiy. Why is chitosan mucoadhesive?, *Biomacromolecules*, **2008**, 9(7), 1837-1842.
19. M. Kurakula, S. Gorityala, K. Moharir. Recent trends in design and evaluation of chitosan-based colon targeted drug delivery systems: Update 2020, *Journal of Drug Delivery Science and Technology*, **2021**, 64, 102579.
20. K. Ling, H. Wu, A. S. Neish, J. A. Champion. Alginate/chitosan microparticles for gastric passage and intestinal release of therapeutic protein nanoparticles, *Journal of Controlled Release*, **2019**, 295, 174-186.
21. M. K. Amin, J. S. Boateng. Enhancing stability and mucoadhesive properties of chitosan nanoparticles by surface modification with sodium alginate and polyethylene glycol for potential oral mucosa vaccine delivery, *Marine Drugs*, **2022**, 20(3), 156.
22. T. Chen, S. Li, W. Zhu, Z. Liang, Q. Zeng. Self-assembly pH-sensitive chitosan/alginate coated polyelectrolyte complexes for oral delivery of insulin, *Journal of Microencapsulation*, **2019**, 36(1), 96-107.
23. L. Tang, Y. H. Chen, Q. Wang, X. H. Wang, Q. X. Wu, Z. F. Ding. Microencapsulation of functional ovalbumin and bovine serum albumin with polylysine-alginate complex for sustained protein vehicle's development, *Food Chemistry*, **2022**, 368, 130902.
24. N. Bock, T. R. Dargaville, M. A. Woodruff. Electrospraying of polymers with therapeutic molecules: state of the art, *Progress in Polymer Science*, **2012**, 37(11), 1510-1551.
25. H. T. T. Nguyen, T. N. Tran, A. C. Ha, P. D. Huynh. Impact of deacetylation degree on properties of chitosan for formation of electrosprayed nanoparticles, *Journal of Nanotechnology*, **2022**, 2288892.
26. L. Zhang, J. Huang, T. Si, R. X. Xu. Coaxial electrospray of microparticles and nanoparticles for biomedical applications, *Expert Review of Medical Devices*, **2012**, 9(6), 595-612.
27. Z. Mai, J. Chen, T. He, Y. Hu, X. Dong, H. Zhang, W. Huang, F. Ko, W. Zhou. Electrospray biodegradable microcapsules loaded with curcumin for drug delivery

- systems with high bioactivity, *RSC Advances*, **2017**, 7(3), 1724-1734 .
28. C. H. Zheng, W. Q. Liang, F. Li, Y. P. Zhang, W. J. Fang. Optimization and characterization of chitosan-coated alginate microcapsules containing albumin, *Die Pharmazie-An International Journal of Pharmaceutical Sciences*, **2005**, 60(6), 434-438.
 29. N. J. Kruger. The Bradford method for protein quantitation. *The protein protocols handbook*, Springer Protocols Handbooks, Humana Press, Totowa, 2009.
 30. B. W. Lee, J. H. Ha, H.G. Shin, S. H. Jeong, J. H. Kim, J. Lee, J. Y. Park, H. J. Kwon, K. Jung, W. S. Lee. Linderia obtusiloba attenuates oxidative stress and airway inflammation in a murine model of ovalbumin-challenged asthma, *Antioxidants*, **2020**, 9(7), 563.
 31. L. Zhang, J. Huang, T. Si, R. X. Xu. Coaxial electrospray of microparticles and nanoparticles for biomedical applications, *Expert Review of Medical Devices*, **2012**, 9(6), 595-612.
 32. P. Mukhopadhyay, S. Chakraborty, S. Bhattacharya, R. Mishra, P. P. Kundu. pH-sensitive chitosan/alginate core-shell nanoparticles for efficient and safe oral insulin delivery, *International Journal of Biological Macromolecules*, **2015**, 72, 640-648.
 33. X. Chen, L. Jia, X. Yin, J. Cheng, J. Lu. Spraying modes in coaxial jet electrospray with outer driving liquid, *Physics of Fluids*, **2005**, 17(3), 032101.
 34. J. F. Hu, S. F. Li, G. R. Nair, W. T. Wu. Predicting chitosan particle size produced by electrohydrodynamic atomization, *Chemical Engineering Science*, **2012**, 82, 159-165.
 35. A. Sosnik. Production of drug-loaded polymeric nanoparticles by electrospraying technology, *Journal of Biomedical Nanotechnology*, **2014**, 10(9), 2200-2217.
 36. S. Ghayempour, S. M. Mortazavi. Fabrication of micro-nanocapsules by a new electrospraying method using coaxial jets and examination of effective parameters on their production, *Journal of Electrostatics*, **2013**, 71(4), 717-727.
 37. C. Chen, W. Liu, P. Jiang, T. Hong. Coaxial electrohydrodynamic atomization for the production of drug-loaded micro/nanoparticles, *Micromachines*, **2019**, 10(2), 125.
 38. P. Cao, F. Y. Han, L. Grøndahl, Z. P. Xu, L. Li. Enhanced oral vaccine efficacy of polysaccharide-coated calcium phosphate nanoparticles, *ACS Omega*, **2020**, 5(29), 18185-18197.
 39. M. C. G. Guillén, M. P. Montero. Enhancement of oral bioavailability of natural compounds and probiotics by mucoadhesive tailored biopolymer-based nanoparticles: a review, *Food Hydrocolloids*, **2021**, 118, 106772.
 40. J. S. Crater, R. L. Carrier. Barrier properties of gastrointestinal mucus to nanoparticle transport, *Macromolecular Bioscience*, **2010**, 10(12), 1473-1483.
 41. P. I. Soares, A. I. Sousa, J. C. Silva, I. M. Ferreira, C. M. Novo, J. P. Borges. Chitosan-based nanoparticles as drug delivery systems for doxorubicin: optimization and modelling, *Carbohydrate Polymers*, **2016**, 147, 304-312.
 42. P. Kunjumon, G. Viswanathan, S. Baby. Biocompatible madecassoside encapsulated alginate chitosan nanoparticles, their anti-proliferative activity on C6 glioma cells, *Carbohydrate Polymer Technologies and Applications*, **2021**, 2, 100106.
 43. B. Sarmento, D. Ferreira, F. Veiga, A. Ribeiro. Characterization of insulin-loaded alginate nanoparticles produced by ionotropic pre-gelation through DSC and FTIR studies, *Carbohydrate Polymers*, **2006**, 66(1), 1-7.
 44. D. Kulig, A. Z. Korzycka, A. Jarmoluk, K. Marycz. Study on alginate-chitosan complex formed with different polymers ratio, *Polymers*, **2016**, 8(5), 167.
 45. L. N. H. Cao, T. V. Nguyen, N. Q. Nguyen, T. B. T. Nguyen, H. V. T. Luong, D. T. Pham.

- Alginate functionalized chitosan nanoparticles using multilayer co-axial electro-spraying for ovalbumin controlled release via oral delivery, *Journal of Drug Delivery Science and Technology*, **2024**, 96, 105733.
46. C. Y. Karakas, H. R. Ordu, F. Bozkurt, A. Karadag. Electrosprayed chitosan-coated alginate–pectin beads as potential system for colon-targeted delivery of ellagic acid, *Journal of the Science of Food and Agriculture*, **2022**, 102(3), 965-975.
 47. M. Chatterjee, N. Jaiswal, A. Hens, N. Mahata, N. Chanda. Development of 6-thioguanine conjugated PLGA nanoparticles through thioester bond formation: benefits of electrospray mediated drug encapsulation and sustained release in cancer therapeutic applications, *Materials Science and Engineering: C*, **2020**, 114, 111029.
 48. D. Zaeim, M. S. Jamab, B. Ghorani, R. Kadkhodae, R. H. Tromp. Electrospray assisted fabrication of hydrogel microcapsules by single-and double-stage procedures for encapsulation of probiotics, *Food and Bioproducts Processing*, **2017**, 102, 250-259.
 49. J. M. Unagolla, A. C. Jayasuriya. Drug transport mechanisms and in vitro release kinetics of vancomycin encapsulated chitosan-alginate polyelectrolyte microparticles as a controlled drug delivery system, *European Journal of Pharmaceutical Sciences*, **2018**, 114, 199-209.
 50. X. Sun, C. Liu, A. M. Omer, L. Y. Yang, X. K. Ouyang. Dual-layered pH-sensitive alginate/chitosan/kappa-carrageenan microbeads for colon-targeted release of 5-fluorouracil, *International Journal of Biological Macromolecules*, **2019**, 132, 487-494.
 51. M. Ge, Y. Li, C. Zhu, G. Liang, J. Alam, G. Hu, Y. Gui, J. Rashid. Preparation of organic-modified magadiite–magnetic nanocomposite particles as an effective nanohybrid drug carrier material for cancer treatment and its properties of sustained release mechanism by Korsmeyer–Peppas kinetic model, *Journal of Materials Science*, **2021**, 56(25), 14270-14286.



© 2025 by the authors. This Open Access Article is licensed under the Creative Commons Attribution-NonCommercial 4.0 International (CC BY-NC 4.0) license (<https://creativecommons.org/licenses/by-nc/4.0/>).

Các mô hình mạng nơ ron hồi quy dùng cho dự báo điện gió

Nguyễn Văn Ninh, Nguyễn Xuân Thâm, Thân Văn Thông,
Đoàn Thanh Bảo, Lương Ngọc Toàn, Lê Tuấn Hộ*

Khoa Kỹ thuật và Công nghệ, Trường Đại học Quy Nhơn, Việt Nam

*Ngày nhận bài: 14/04/2025; Ngày sửa bài: 06/08/2025;
Ngày nhận đăng: 12/08/2025; Ngày xuất bản: 28/10/2025*

TÓM TẮT

Các giá trị lịch sử của công suất phát điện gió thường được sử dụng trong hầu hết các mô hình và phương pháp dự báo điện gió trong các tài liệu. Tuy nhiên, các thông số vận hành có thể ảnh hưởng đến kết quả dự báo chẳng hạn như tốc độ gió, góc pitch, nhiệt độ môi trường, vị trí nacelle, và hướng gió chưa được xem xét trong các phương pháp hiện tại. Do đó, mục tiêu cơ bản của bài báo này là đề xuất các mô hình mạng nơ ron hồi quy dùng cho dự báo công suất phát điện gió có xem xét các tham số vận hành này. Nghiên cứu này xem xét dữ liệu công suất phát điện gió và các tham số vận hành tương ứng từ máy phát tuabin gió số 5 của 1 trang trại gió. Dữ liệu từ ngày 01 tháng 7 năm 2024 đến 31 tháng 7 năm 2024 được thu thập từ hệ thống SCADA. Đầu tiên, mô hình mạng nơ ron tự hồi quy phi tuyến có các đầu vào được áp dụng để dự báo công suất phát điện gió. Thứ hai, mô hình mạng hồi quy lớp được sử dụng để dự báo công suất phát điện gió. Thứ ba, mô hình mạng nơ ron trễ phân tán được dùng để dự báo công suất phát điện gió. Thứ tư, mô hình mạng nơ ron trễ thời gian được huấn luyện để ước lượng điện gió. Cuối cùng, các mô hình mạng nơ ron hồi quy này được so sánh để xác định mô hình dự báo công suất phát điện gió tốt hơn khi xét theo các tiêu chí sai số tuyệt đối trung bình, sai số phần trăm tuyệt đối trung bình và sai số bình phương trung bình.

Từ khóa: *Dự báo điện gió, mạng nơ ron tự hồi quy phi tuyến có các đầu vào, mạng nơ ron hồi quy lớp, mạng nơ ron trễ phân tán, mạng nơ ron trễ thời gian.*

*Tác giả liên hệ chính.

Email: tuanhole@qnu.edu.vn

Recurrent neural network models for wind power forecasting

Van Ninh Nguyen, Xuan Tham Nguyen, Van Thong Than, Thanh Bao Doan,
Ngoc Toan Luong, Tuan Ho Le*

Faculty of Engineering and Technology, Quy Nhon University, Vietnam

Received: 14/04/2025; Revised: 06/08/2025;

Accepted: 12/08/2025; Published: 28/10/2025

ABSTRACT

The historical values of wind power generation are generally utilized in most forecasting models and methods in the literature. Unfortunately, the operational parameters such as wind speed, pitch angle, ambient temperature, nacelle position, and wind direction may affect the forecasting results. Therefore, the primary objective of this paper is to propose recurrent neural network models for wind power generation forecasting considering these operational parameters. In this study, the wind power generation data and the associated operational parameters from the wind turbine generator 05 of a wind farm are investigated. The data from July 1st, 2024 to July 31st, 2024 is collected from the SCADA system. Firstly, the nonlinear autoregressive neural network with external input is applied to make the wind power generation prediction. Secondly, the layer recurrent neural network model is employed to forecast wind power generation. Thirdly, the distributed delay neural network model is implemented to predict wind power generation. Fourthly, the time delay neural network model is trained to estimate the wind power. Finally, these recurrent neural network models are compared to determine the better wind power generation forecasting model in terms of mean absolute error, mean absolute percent error, and root mean square error.

Keywords: *Wind power forecasting, nonlinear autoregressive neural network with external input, layer recurrent neural network, distributed delay neural network, time delay neural network.*

1. INTRODUCTION

The rapid increase in energy demand has driven the search for alternative energy sources, in addition to traditional ones that are depleting and causing pollution issues. Wind power is a clean and renewable source. According to the Global Wind Report 2024 (by Global Wind Energy Council), it is shown that 2023 recorded the highest number of new installations in history for onshore wind (over 100 GW) and the second highest for offshore wind (11 GW). Wind energy installations will increase from a level of 117 GW in 2023 to at least 320 GW of annual

installations by 2030. Actually, wind power generation plays a significant role in electricity supply. However, wind energy integration into power systems presents inherent unpredictability because of the intermittent nature of wind energy.¹ As wind energy makes significant penetration into the electricity grid, the need for accurate predictions of wind power generation becomes critical and urgent.^{2,3} To address these challenges, wind power forecasting (WPF) has emerged as a valuable solution. Consequently, numerous WPF models and methods have been proposed and implemented in the literature.

*Corresponding author.

Email: tuanhole@qnu.edu.vn

Based on the forecasting time horizon, WPF can be categorized into ultra short-term, short-term, medium-term, and long-term. Various types of forecasting models and methods are developed for wind power generation time series. The traditional statistical models and methods are usually applied by using the previous historical data to perform a forecast. In WPF, the statistical models are commonly applied as exponential smoothing approach,^{4,5} autoregressive,⁶ autoregressive moving average (ARMA),^{7,8} autoregressive integrated moving average (ARIMA).^{9,10} Statistical models are more user-friendly and cost-effective to develop than other types of models. Statistical methods primarily rely on historical wind data to forecast the upcoming few hours, making them suitable for short-term predictions. While statistical forecasting models such as ARIMA and exponential smoothing are simple and interpretable, they suffer from limitations such as linearity assumptions, difficulty incorporating multiple external variables, and poor scalability to nonlinear or high-dimensional problems. Besides, artificial intelligence (AI)-based models are another research direction in WPF. Fundamentally, AI-based models, when adequately trained, have the potential to outperform traditional statistical models in forecasting accuracy. Depending on the structure of neural networks (NNs), various models are proposed in wind power generation forecasting. Feed-forward NN models are used to make the wind power prediction such as multi-layer perceptron¹¹ and back-propagation (BP) NN.¹² Another kind of NN model with feedback known as the recurrent NN (RNN) model is also widely applied in this research direction. The RNN model-based WPF methods are Elman NN,¹³ layer RNN,¹⁴ nonlinear autoregressive NN,¹⁵ long short-term memory (LSTM),¹⁶ bidirectional LSTM,¹⁷ gated recurrent unit,¹⁸ and echo state network.¹⁹ Deep learning models such as LSTM, bidirectional LSTM, gated recurrent unit, and echo state network have demonstrated improved forecasting performance compared to earlier NN architectures. Moreover,

support vector machine,²⁰ gradient boosting regression tree algorithms,²¹ and ensemble model²² belonging to machine learning are also implemented in WPF. These machine learning models have also been shown to outperform NN models in specific research cases. Other attempts have been made to combine different models or methods in various ways to improve the forecasting results. These include combinations such as autoregressive fractionally integrated moving average and least square support vector machine,²² boosting algorithm and ARMA model,²³ hybrid CEEMDAN-EWT deep learning method,²⁴ and neuro wavelet and LSTM models.²⁵ These combined models have also demonstrated improved forecasting performance compared to each individual model. In these existing studies, most WPF models address the wind power generation time series data issues without taking the operational parameters into account. These operating parameters have a direct impact on the power output of wind turbines in real-world conditions; therefore, they influence the accuracy of forecasting results. Some significant operational parameters can be considered wind speed, pitch angle, ambient temperature, nacelle position, and wind direction. Therefore, in this paper, one of the six turbines of a wind farm, located on the south-central coast, Vietnam with a 114-metre height and a 132-metre rotor is considered. These operational parameters and wind power generation time series data are collected at 10-minute intervals from the SCADA system. The dataset spans from July 01st, 2024 to July 31st, 2024. This data is divided into three different case studies. Subsequently, numerous RNN models are proposed for wind power forecasting, including nonlinear autoregressive NN with external input (Narxnet), layer RNN (Layrecnet), distributed delay NN (Distdelaynet), and time delay neural network (Timedelaynet). These models incorporate both operational parameters and wind power generation data. To evaluate the performance of WPF models, evaluation criteria such as mean absolute error (MAE), mean absolute

percent error (MAPE), and root mean square error (RMSE) are employed. Based on the final results, the most effective forecasting model can be determined. An overview of wind power generation forecasting using RNN models can be represented in Figure 1.

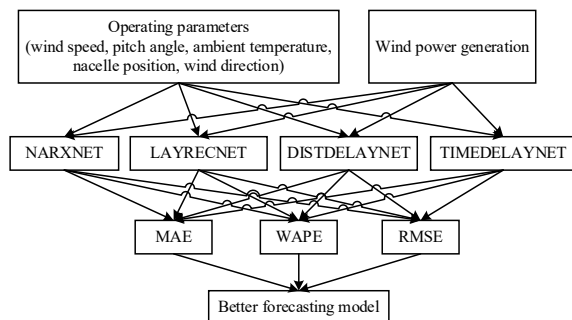


Figure 1. Overview of wind power generation forecasting using RNN models.

2. METHODOLOGY

2.1. Nonlinear autoregressive neural network with external input

Narxnet is a nonlinear autoregressive model with exogenous inputs commonly used in time series modeling. Narxnet is first formally proposed and popularized in the mid-1990s by Lin *et al.*²⁶ In this model, the current value of a time series depends on both past values of the same series and current and past values of exogenous series. The Narxnet model, applied to time series forecasting, can be mathematically represented as follows:

$$y_t = f(y_{t-1}, y_{t-2}, \dots, x_t, x_{t-1}, x_{t-2}, \dots) \quad (1)$$

where f denotes an unknown nonlinear function (i.e., transfer function or activation function), y_t denotes the predicted value of the time series data of y at a discrete time t , and x_t denotes the externally determined variable. The Narxnet model can be illustrated in Figure 2.

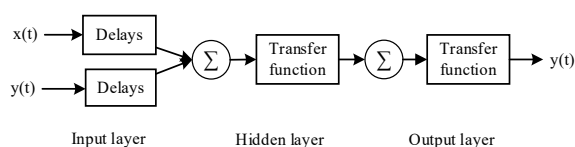


Figure 2. Narxnet model.

2.2. Layer recurrent neural network

Layrecnet is another type of RNN models. The concept of Layrecnet model is proposed by Schmidhuber.²⁷ In this architecture, the output of the hidden layer is fed back to the input layer with delays. As a result, the network is capable of exhibiting an infinite dynamic response to time series input data. The Layrecnet model can be illustrated in Figure 3.

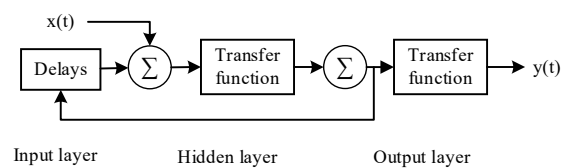


Figure 3. Layrecnet model.

2.3. Distributed delay neural network

Distdelaynet is another type of RNN model. The concept of Distdelaynet model is proposed by Waibel *et al.*²⁸ The input and hidden layers have a tap delay line associated with them. Therefore, the network exhibits a finite dynamic response to time series input data. The Distdelaynet model can be illustrated in Figure 4.

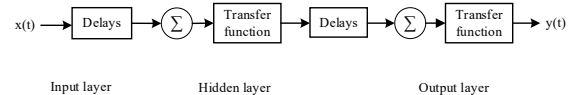


Figure 4. Distdelaynet model.

2.4. Time delay neural network

Timedelaynet is another type of RNN model. The concept of Distdelaynet model is also proposed by Waibel *et al.*²⁸ In this NN model, the input layer has a tap delay line associated with it. Therefore, the network exhibits a finite dynamic response to time series input data. The Timedelaynet model can be illustrated in Figure 5.

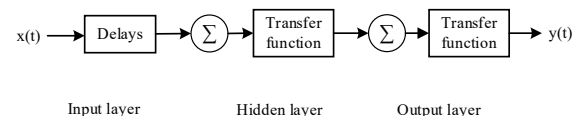


Figure 5. Timedelaynet model.

2.5. Transfer function

In NN models, transfer functions (also known

as activation functions) play a crucial role in introducing nonlinearity and determining the output of each neuron. Commonly, three types of transfer functions are employed, each serving a specific purpose within the network architecture:

Linear (i.e., purelin):

$$f = x. \quad (1)$$

Hyperbolic tangent sigmoid (i.e., tansig):

$$f = \frac{2}{1 + e^{-2x}} - 1. \quad (2)$$

Log-sigmoid (i.e., logsig):

$$f = \frac{1}{1 + e^{-x}}. \quad (3)$$

2.6. Training algorithm

The training process of a NN begins with the random initialization of connection weights, which serve as the adjustable parameters controlling how input signals are transformed through the network layers. These weights are then iteratively updated to minimize a predefined cost (or loss) function, which quantifies the discrepancy between the network's predicted output and the actual target value.²⁹ This minimization is typically performed using gradient-based optimization algorithms that adjust the weights in the direction that reduces the error.

Several well-established training algorithms are employed for this purpose, each with distinct convergence characteristics and computational requirements:

- Resilient BP (i.e., trainrp): Focuses on the sign of the gradient rather than its magnitude to achieve stable convergence, especially in noisy or flat error surfaces.
- Bayesian regularization BP (i.e., trainbr): Incorporates a regularization term into the cost function to prevent overfitting by balancing model complexity and data fitting.
- BFGS quasi-Newton BP (i.e., trainbfg): Utilizes second-order derivative approximations to speed up convergence, particularly effective in medium-sized networks.

➤ Levenberg–Marquardt BP (i.e., trainlm): Combines the advantages of the Gauss–Newton algorithm and gradient descent, offering fast convergence and high accuracy for smaller datasets or networks.

The choice of training algorithm can significantly affect the learning efficiency, generalization ability, and computational cost of the neural network model.

2.7. Evaluation criteria

In this study, three evaluation criteria are employed to assess the performance of the forecasting models. These criteria are widely adopted in time series forecasting tasks, as they provide comprehensive insights into prediction accuracy and error distribution. Specifically, the evaluation metrics used are as follows:

MAE:

$$MAE = \frac{1}{n} \sum_{t=1}^n |A_t - F_t| \quad (4)$$

where A_t is the actual value, F_t is the forecasted value, and n is the number of observations.

WAPE:

$$WAPE = \frac{\sum_{t=1}^n |A_t - F_t|}{\sum_{t=1}^n |A_t|}. \quad (5)$$

RMSE:

$$RMSE = \sqrt{\frac{1}{n} \sum_{t=1}^n (A_t - F_t)^2}. \quad (6)$$

3. CASE STUDIES

The time-series data from the wind turbine no. 05 (3.5 MW, 114-meter hub height, 132-meter rotor diameter) in a wind farm were collected from July 01st, 2024 to July 31st, 2024 (i.e., case study 3) with 10-minute intervals. Two investigated sub-periods during this timeframe are from July 01st, 2024 to July 07th, 2024 (i.e., case study 1) and from July 25th, 2024 to July 31st, 2024 (i.e., case study 2). The total number of observations in the three case studies is provided in Table 1.

Table 1. Number of observations.

Data	Case study 1	Case study 2	Case study 3
Number of observations	1008	1008	4464

The wind speed, pitch angle, ambient temperature, nacelle position, and wind direction from July 01st, 2024 to July 31st, 2024 are illustrated in Figures 6, 7, 8, 9, and 10, respectively.

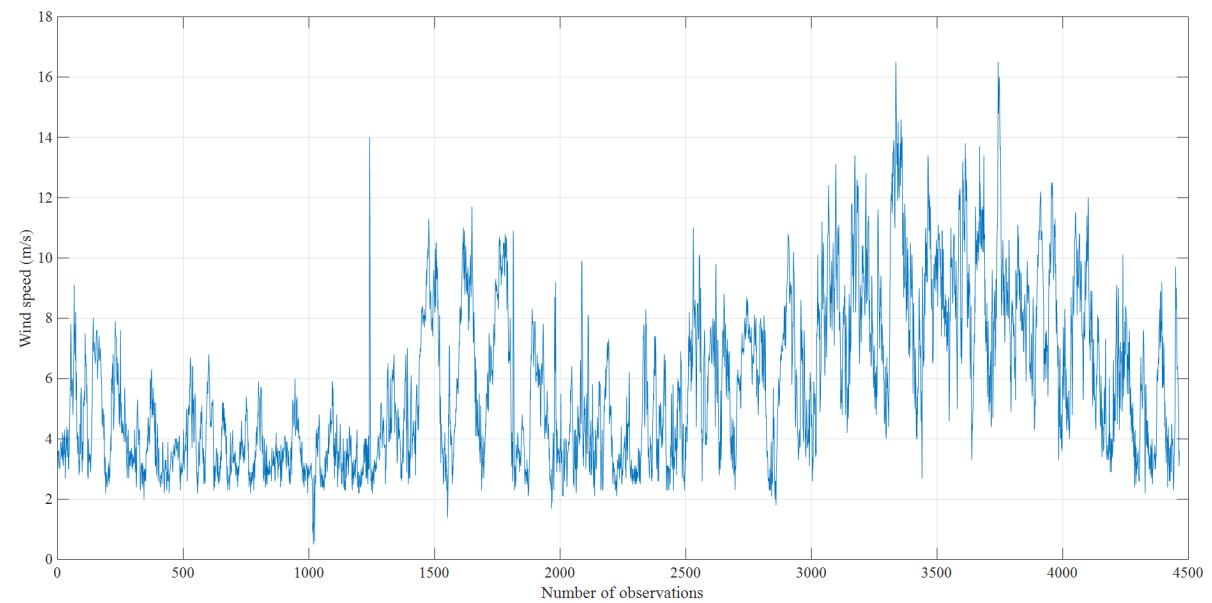


Figure 6. Wind speed.

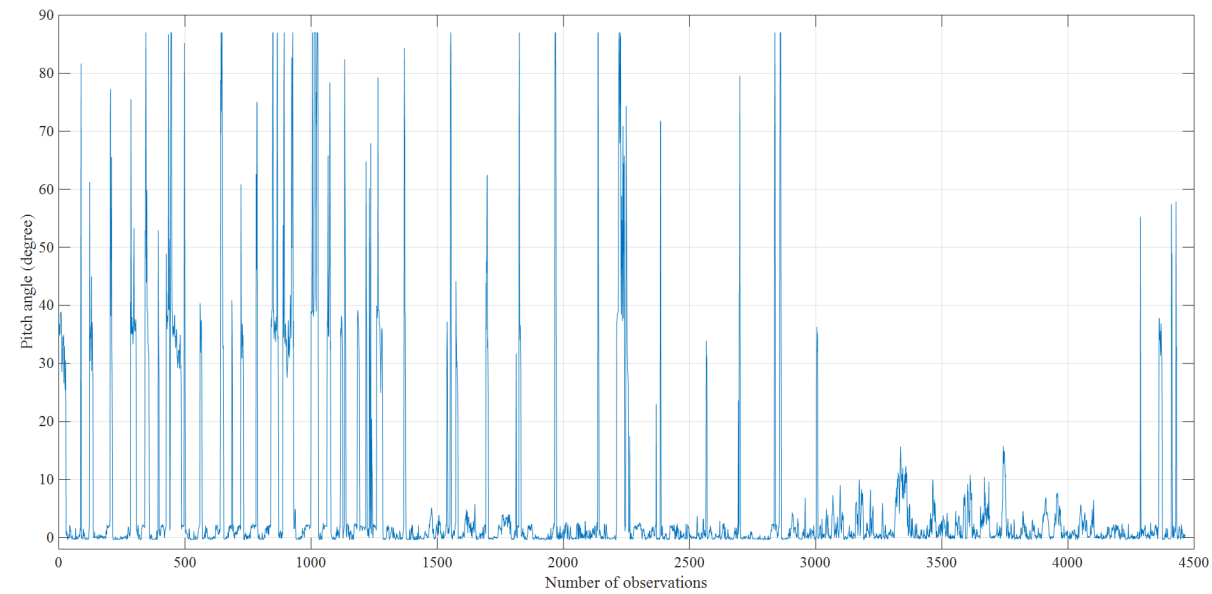


Figure 7. Pitch angle.

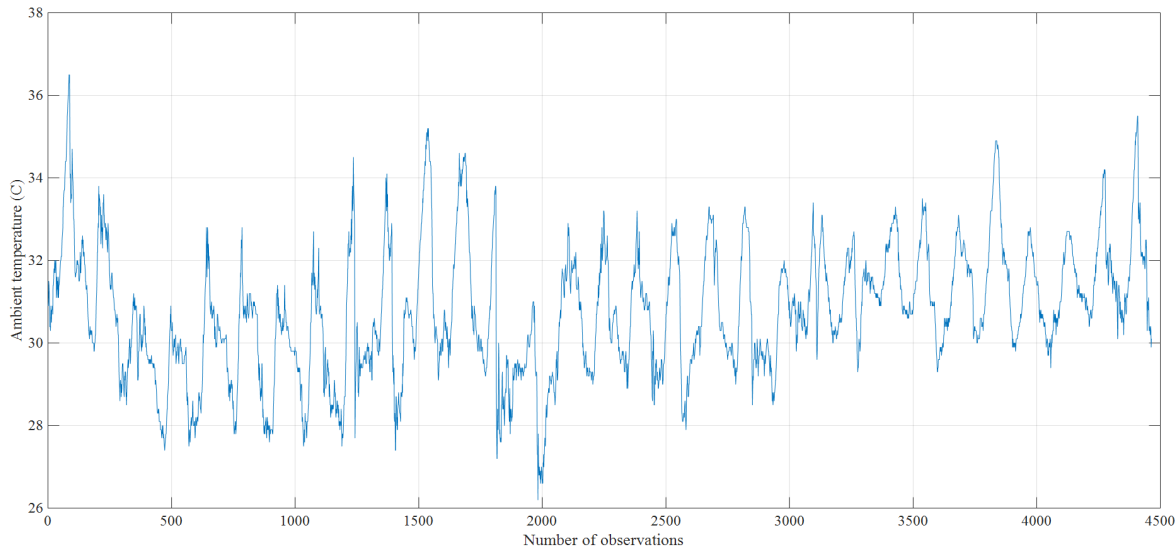


Figure 8. Ambient temperature.

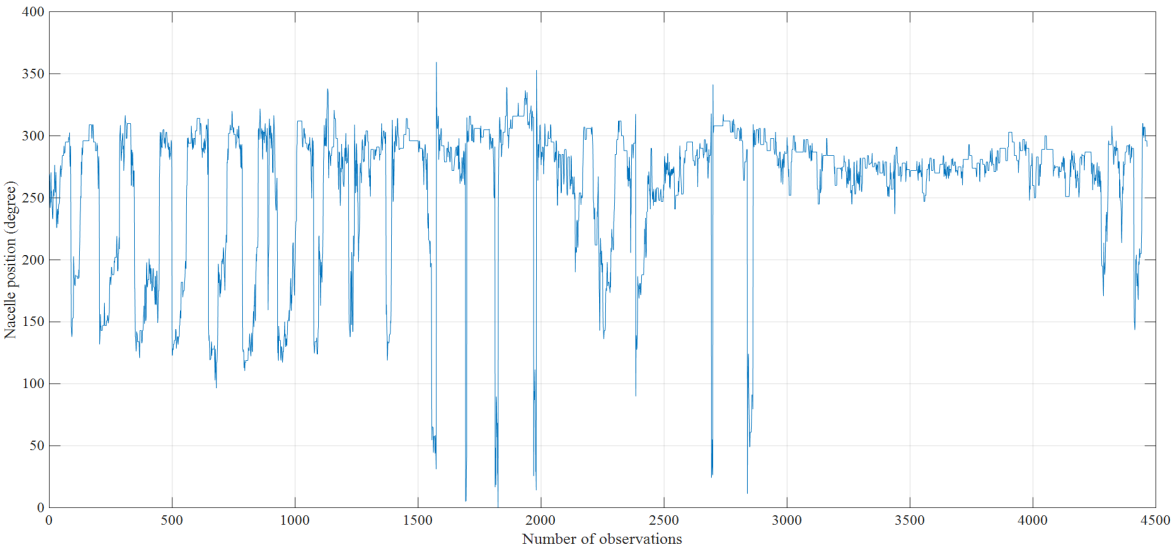


Figure 9. Nacelle position.

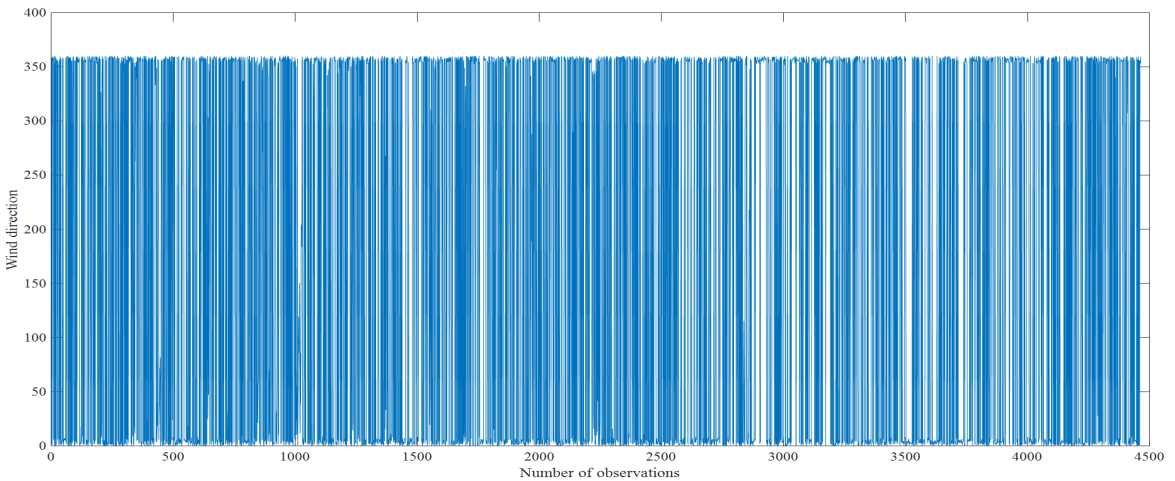


Figure 10. Wind direction.

All the recurrent NN models with one hidden layer have been implemented. Moreover, these models, each configured with one feedback delay in their respective structures, are coded and trained in Matlab software. The transfer function, training algorithms, and number of hidden neurons of all the trained Narxnet, Layrecnet, Distdelaynet, and Timedelaynet models for case studies 1, 2, and 3 are represented in Tables 2, 3, and 4, respectively.

Table 2. Information of the recurrent NN models in case study 1.

Model	Transfer function	Training algorithm	Hidden neurons
Narxnet	Tansig	Trainbr	47
Layrecnet	Tansig	Trainbr	30
Distdelaynet	Tansig	Trainbr	143
Timedelaynet	Logsig	Trainbr	253

Table 3. Information of the recurrent NN models in case study 2.

Model	Transfer function	Training algorithm	Hidden neurons
Narxnet	Logsig	Trainbr	21
Layrecnet	Tansig	Trainbr	79
Distdelaynet	Tansig	Trainlm	232
Timedelaynet	Tansig	Trainbr	301

Table 4. Information of the recurrent NN models in case study 3.

Model	Transfer function	Training algorithm	Hidden neurons
Narxnet	Tansig	Trainlm	32
Layrecnet	Tansig	Trainbr	48
Distdelaynet	Logsig	Trainlm	372
Timedelaynet	Tansig	Trainbr	180

The performance evaluation results of all RNN forecasting models across the three case studies are presented in Tables 5, 6, and 7, respectively. These tables provide a comprehensive comparison based on multiple error metrics, allowing for a thorough assessment of each model's forecasting accuracy and consistency under varying data conditions. By analyzing these evaluation values, the relative effectiveness of each RNN architecture can be quantitatively compared across different time periods.

Table 5. Evaluation criteria of the proposed forecasting models in case study 1.

Model	MAE	WAPE(%)	RMSE
Narxnet	56.0914	22.3584	87.4324
Layrecnet	18.6198	7.4220	24.9867
Distdelaynet	65.9784	26.2994	130.9846
Timedelaynet	49.9778	19.9215	98.3547

Table 6. Evaluation criteria of the proposed forecasting models in case study 2.

Model	MAE	WAPE(%)	RMSE
Narxnet	269.7987	17.0885	382.9613
Layrecnet	80.9957	5.1301	104.7831
Distdelaynet	341.8460	21.6519	490.9236
Timedelaynet	207.4276	13.1381	289.5628

Table 7. Evaluation criteria of the proposed forecasting models in case study 3.

Model	MAE	WAPE(%)	RMSE
Narxnet	168.6443	18.3242	263.3885
Layrecnet	41.3159	4.4892	58.1676
Distdelaynet	248.0907	26.9565	388.8692
Timedelaynet	164.6041	17.8852	256.3703

Based on the results presented in Table 5, the Narxnet model yields improved forecasting performance over the Distdelaynet model, with lower error values across all three metrics (MAE = 56.0914, WAPE = 22.3584%, RMSE = 87.4324 vs. MAE = 65.9784, WAPE = 26.2994%, RMSE = 130.9846). Furthermore, the Timedelaynet model demonstrates superior predictive accuracy compared to Narxnet, achieving a lower MAE of 49.9778, WAPE of 19.9215%, and RMSE of 98.3547. Among all models, the Layrecnet model achieves the best performance in this case study, with significantly lower errors (MAE = 18.6198, WAPE = 7.4220%, RMSE = 24.9867), indicating its strong capability in capturing the temporal dynamics of wind power data.

Similarly, Table 6 shows that the Layrecnet model (MAE = 80.9957, WAPE = 5.1301%, RMSE = 104.7831) continues to outperform the Timedelaynet (MAE = 207.4276, WAPE = 13.1381%, RMSE = 289.5628), Narxnet (MAE = 269.7987, WAPE = 17.0885%, RMSE = 382.9613),

and Distdelaynet models (MAE = 341.8460, WAPE = 21.6519%, RMSE = 490.9236), further confirming its robustness across different time periods.

In Table 7, the evaluation results also confirm the superiority of the Layrecnet model in the third case study. It achieves the lowest error values (MAE = 41.3159, WAPE = 4.4892%, RMSE = 58.1676), outperforming Timedelaynet (MAE = 164.6041, WAPE = 17.8852%, RMSE = 256.3703), Narxnet (MAE = 168.6443, WAPE = 18.3242%, RMSE = 263.3885), and Distdelaynet (MAE = 248.0907, WAPE = 26.9565%, RMSE = 388.8692).

Figures 11, 12, and 13 illustrate the comparison between the actual wind power generation data and the forecasting results from the four models across case studies 1, 2, and 3, respectively. The actual wind power values are shown as solid blue lines, while the predicted values generated by the Narxnet, Layrecnet, Distdelaynet, and Timedelaynet models are represented by dashed lines in red, blue, purple, and light blue, respectively.

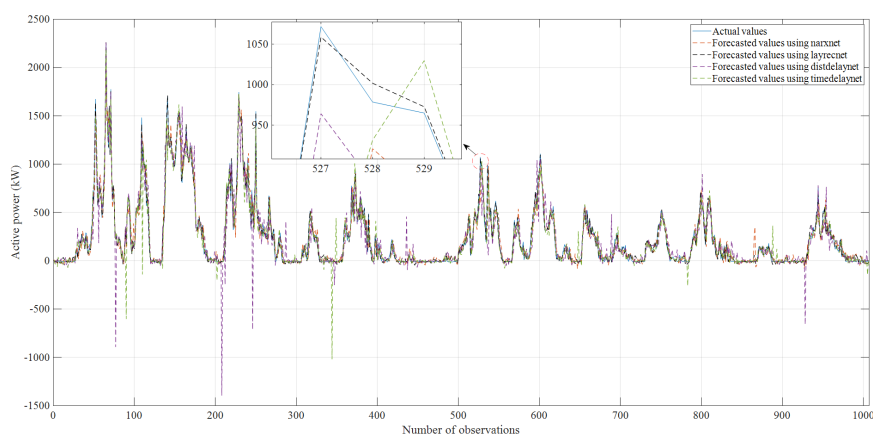


Figure 11. The forecasted results of wind power generation in case study 1.

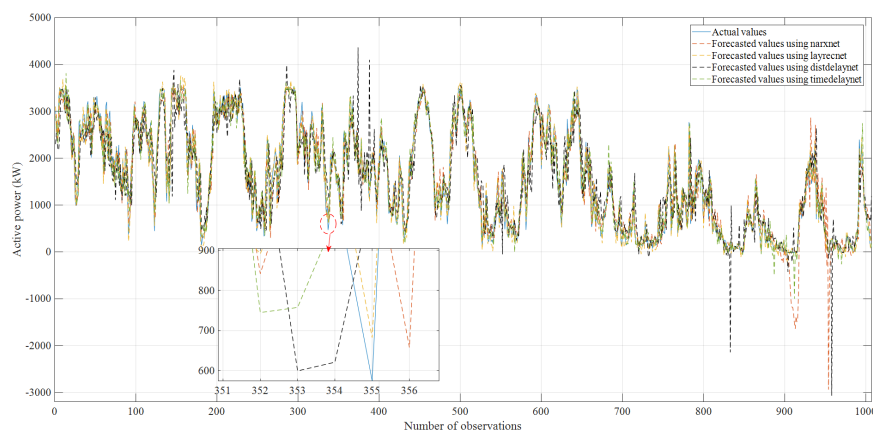


Figure 12. The forecasted results of wind power generation in case study 2.

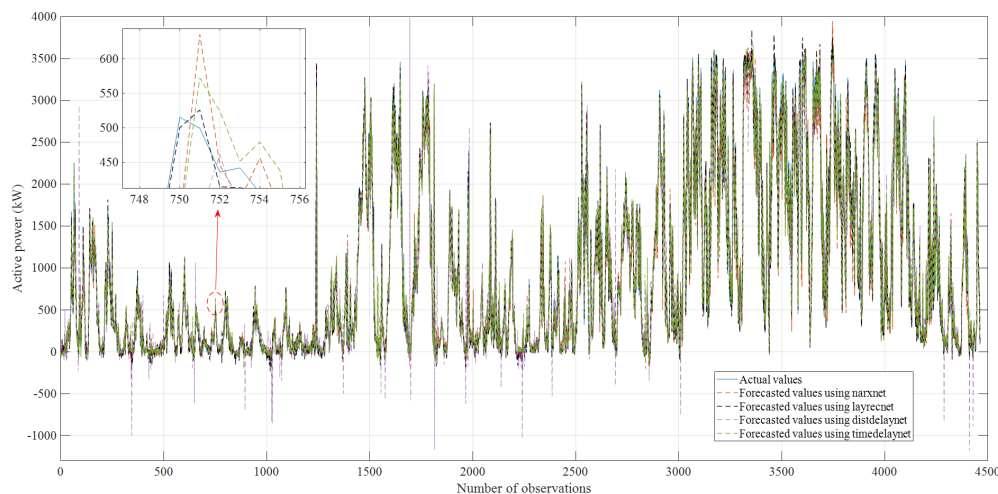


Figure 13. The forecasted results of wind power generation in case study 3.

4. CONCLUSION

In this paper, operational parameters such as wind speed, pitch angle, ambient temperature, nacelle position, and wind direction are taken into account for wind power generation forecasting. A wind farm comprising six turbines, each with a capacity of 3.5 MW, a hub height of 114 meters, and a rotor diameter of 132 meters, is considered. The time series data of turbine no. 05 from July 01st, 2024 to July 31st, 2024 is collected. Several RNN architectures consisting of Narxnet, Layrecnet, Distdelaynet, and Timedelaynet are proposed as alternative approaches for wind power generation forecasting. Among these models, the Layrecnet model demonstrates superior performance WPF results compared to these other models in terms of MAE, WAPE, and RMSE. For further study, the optimal structures of these models can be identified to provide better solutions. Moreover, to improve model reliability, anomalous data points and outliers are identified and filtered out prior to training the forecasting models.

Acknowledgments

This research is conducted within the framework of science and technology projects at institutional level of Quy Nhon University under the project code T2024.847.18.

REFERENCES

1. F. Ullah, X. Zhang, M. Khan, M. S. Mastoi, H. M. Munir, A. Flah, Y. Said. A comprehensive review of wind power integration and energy storage technologies for modern grid frequency regulation, *Heliyon*, **2024**, 10(9), e30466.
2. G. Li, J. Shi. On comparing three artificial neural networks for wind speed forecasting, *Applied Energy*, **2010**, 87(7), 2313-2320.
3. J. Lerner, M. Grundmeyer, M. Garvert. The importance of wind forecasting, *Renewable Energy Focus*, **2009**, 10(2), 64-66.
4. A. Kusiak, Z. Zhang. Short-horizon prediction of wind power: a data-driven approach, *IEEE Transactions on Energy Conversion*, **2010**, 25(4), 1112-1122.
5. X. Zheng, T. Jin. A reliable method of wind power fluctuation smoothing strategy based on multidimensional non-linear exponential smoothing short-term forecasting, *IET Renewable Power Generation*, **2022**, 16(16), 3573-3586.
6. A. Anwar, A. N. Mahmood. Enhanced estimation of Autoregressive wind power prediction model using constriction factor particle swarm optimization, 2014 9th IEEE

- Conference on Industrial Electronics and Applications, Hangzhou, China, 2014.
7. J. Wang, Q. Zhou, X. Zhang. *Wind power forecasting based on time series ARMA model*, IOP Conference Series: Earth and Environmental Science, **2018**, 199(2), 1-6.
 8. R. B. Magadum, S. Bilagi, S. Bhandarkar, A. Patil, A. Joshi. *Short-term wind power forecast using time series analysis: Autoregressive moving-average model (ARMA)*, Recent developments in electrical and electronics engineering: select proceedings of ICRDEEE 2022, Springer Nature Singapore, Singapore, 2023.
 9. F. A. Eldali, T. M. Hansen, S. Suryanarayanan, E. K. P. Chong. *Employing ARIMA models to improve wind power forecasts: a case study in ERCOT*, 2016 North American Power Symposium (NAPS), IEEE, Denver, CO, USA, 2016.
 10. B. M. Hodge, A. Zeiler, D. Brooks, G. Blau, J. Pekny, G. Reklatis. Improved wind power forecasting with ARIMA models, *Computer Aided Chemical Engineering*, **2011**, 29, 1789-1793.
 11. J. Li, H. Cheng, X. Zhou, M. Wang, Z. Wang. *Optimization of multi-layer perceptron for wind power generation prediction based on improved grey wolf algorithm*, International Symposium on New Energy and Electrical Technology, Springer Nature Singapore, Singapore, 2023.
 12. W. Sun, Y. Wang. Short-term wind speed forecasting based on fast ensemble empirical mode decomposition, phase space reconstruction, sample entropy and improved back-propagation neural network, *Energy Conversion and Management*, **2018**, 157, 1-12.
 13. J. Huang, R. Qin. Elman neural network considering dynamic time delay estimation for short-term forecasting of offshore wind power, *Applied Energy*, **2024**, 358, 122671.
 14. Z. O. Olaofe, K. A. Folly. *Wind power estimation using recurrent neural network technique*, IEEE Power and Energy Society Conference and Exposition in Africa: Intelligent Grid Integration of Renewable Energy Resources (PowerAfrica), Johannesburg, South Africa, 2012.
 15. A. Ahmed, M. Khalid. *A nonlinear autoregressive neural network model for short-term wind forecasting*, 2017 9th IEEE-GCC Conference and Exhibition (GCCCE), IEEE, Manama, Bahrain, 2017.
 16. Z. Xiao, F. Tang, M. Wang. Wind power short-term forecasting method based on LSTM and multiple error correction, *Sustainability*, **2023**, 15(4), 3798.
 17. L. Jovanovic, K. Kumpf, N. Bacanin, M. Antonijevic, J. Mani, H. Shaker, M. Zivkovic. *Decomposition aided bidirectional long-short-term memory optimized by hybrid metaheuristic applied for wind power forecasting*, International Conference on Computational Sciences and Sustainable Technologies, Bangalore, India, 2023.
 18. Z. Niu, Z. Yu, W. Tang, Q. Wu, M. Reformat. Wind power forecasting using attention-based gated recurrent unit network, *Energy*, **2020**, 196, 117081.
 19. H. Wang, Z. Lei, Y. Liu, J. Peng, J. Liu. Echo state network based ensemble approach for wind power forecasting, *Energy Conversion and Management*, **2019**, 201, 112188.
 20. J. Zeng, W. Qiao. *Support vector machine-based short-term wind power forecasting*, 2011 IEEE/PES Power Systems Conference and Exposition, IEEE, Phoenix, AZ, USA, 2011.
 21. S. Park, S. Jung, J. Lee, J. Hur. A short-term forecasting of wind power outputs based on gradient boosting regression tree algorithms, *Energies*, **2023**, 16(3), 1132.
 22. D. Kumar, R. Abhinav, N. Pindoriya. *An ensemble model for short-term wind power*

- forecasting using deep learning and gradient boosting algorithms*, 2020 21st National Power Systems Conference (NPSC), IEEE, Gandhinagar, India, 2020.
23. X. Yuan, Q. Tan, X. Lei, Y. Yuan, X. Wu. Wind power prediction using hybrid autoregressive fractionally integrated moving average and least square support vector machine, *Energy*, **2017**, 129, 122-137.
 24. Y. Jiang, X. Chen, K. Yu, Y. Liao. Short-term wind power forecasting using hybrid method based on enhanced boosting algorithm, *Journal of Modern Power Systems and Clean Energy*, **2017**, 5(1), 126-133.
 25. H. H. Aly. *A proposed hybrid deep learning model for wind power forecasting*, 2024 ASU International Conference in Emerging Technologies for Sustainability and Intelligent Systems (ICETISIS), IEEE, Manama, Bahrain, 2024.
 26. T. Lin, B. G. Horne, P. Tino, C. L. Giles. Learning long-term dependencies in NARX recurrent neural networks, *IEEE Transactions on Neural Networks*, **1996**, 7(6), 1329-1338.
 27. J. Schmidhuber. Learning complex, extended sequences using the principle of history compression, *Neural Computation*, **1992**, 4(2), 234-242.
 28. A. Waibel, T. Hanazawa, G. Hinton, K. Shikano, K. J. Lang. Phoneme recognition using time-delay neural networks, *IEEE Transactions Acoustics, Speech, Signal Processing*, **1989**, 37(3), 328–339.
 29. T. H. Le, L. Dai, H. Jang, S. Shin. Robust process parameter design methodology: a new estimation approach by using feed-forward neural network structures and machine learning algorithms, *Applied Sciences*, **2022**, 12(6), 2904.



© 2025 by the authors. This Open Access Article is licensed under the Creative Commons Attribution-NonCommercial 4.0 International (CC BY-NC 4.0) license (<https://creativecommons.org/licenses/by-nc/4.0/>).

Nghiên cứu ảnh hưởng của bánh men thuần chủng đến chất lượng sản phẩm cơm rượu

Lê Bích Tuyên, Huỳnh Văn Quốc Cảnh*, Trương Thị Tú Trân

Khoa Khoa học thực phẩm và sức khỏe, Trường Đại học Kiên Giang, Việt Nam

*Ngày nhận bài: 03/06/2025; Ngày sửa bài: 30/07/2025;
Ngày nhận đăng: 12/08/2025; Ngày xuất bản: 28/10/2025*

TÓM TẮT

Nghiên cứu này tập trung sử dụng các chủng vi sinh vật thuần chủng cho vào bánh men nhằm cải tiến chất lượng sản phẩm cơm rượu tại huyện Gò Quao, tỉnh Kiên Giang. Bánh men chứa các vi sinh vật quan trọng như nấm men, nấm mốc và vi khuẩn lactic, đóng vai trò quyết định trong quá trình lên men chuyển hóa tinh bột thành ethanol và tạo hương vị đặc trưng. Qua khảo nghiệm với các nồng độ bánh men thuần chủng (0,4%, 0,6%, 0,8%), kết quả cho thấy nồng độ 0,4% mang lại sản phẩm cơm rượu có chất lượng cảm quan tốt nhất, đồng thời duy trì các chỉ tiêu hóa lý như độ ẩm, pH, và hàm lượng acid lactic ở mức phù hợp, hàm lượng ethanol cao (khoảng 10% v/v). Ngoài ra, nghiên cứu còn khảo sát thời gian lên men trong vòng 2 đến 4 ngày, cho thấy quá trình lên men tối ưu trong 3 ngày, khi các chỉ tiêu như pH, độ cồn và acid lactic ổn định, tạo môi trường lên men lý tưởng đồng thời đảm bảo chất lượng sản phẩm, đồng thời không phát hiện các chất gây hại như methanol và furfural. Nghiên cứu góp phần xây dựng cơ sở khoa học cho việc ứng dụng bánh men thuần chủng, nâng cao tính ổn định và chất lượng cơm rượu truyền thống, đồng thời hỗ trợ bảo tồn và phát triển nghề làm bánh men truyền thống địa phương.

Từ khóa: *Cơm rượu, Gò Quao, nấm men, nấm mốc, vi khuẩn lactic.*

**Tác giả liên hệ chính.*

Email: hvqcanh@vnkgu.edu.vn

Study on the effect of pure fermentation starters (banh men) on the quality of fermented glutinous rice (com ruou)

Le Bich Tuyen, Huynh Van Quoc Canh*, Truong Thi Tu Tran

Faculty of Food Science and Health, Kien Giang University, Vietnam

Received: 03/06/2025; Revised: 30/07/2025;

Accepted: 12/08/2025; Published: 28/10/2025

ABSTRACT

This study aimed to evaluate the impact of using pure microbial strains in fermentation starter (banh men) on the sensory, physicochemical, and microbiological quality of traditional fermented glutinous rice (com ruou) produced in Go Quao District, Kien Giang Province. Traditional banh men harbors essential microorganisms, including yeasts, molds, and lactic acid bacteria, which are responsible for the conversion of starch into alcohol and the development of characteristic flavors during fermentation. The study was conducted using pure fermentation starter (banh men) at concentrations of 0.4%, 0.6%, and 0.8%, with 0.4% yielding the most favorable sensory attributes while maintaining desirable physicochemical parameters such as moisture content, pH, lactic acid levels, and a high alcohol concentration (10.2% v/v). Fermentation time was also evaluated over a period of 2 to 4 days, with results indicating that optimal fermentation occurred after 3 days, when key indicators such as pH, alcohol, and lactic acid stabilized. Importantly, no harmful by-products such as methanol or furfural were detected. This study contributes to establishing a scientific basis for the application of pure fermentation starter in order to improve the stability and quality of traditional fermented glutinous rice (com ruou), while also supporting the preservation and development of the local starter culture production craft.

Keywords: *Fermented glutinous rice, Go Quao, yeast, mold, lactic acid bacteria.*

1. INTRODUCTION

Fermented foods constitute an important part of the culinary culture of many countries, including Vietnam. Among them, traditional fermented glutinous rice (com ruou) is a representative product closely associated with the daily life of the people. The production process of traditional fermented glutinous rice is influenced by starter cakes, which contain molds, yeasts, and lactic acid bacteria. These microorganisms initiate and sustain the transformation of starch into alcohol, while also contributing to the characteristic flavor of traditional fermented glutinous rice.

Starter cakes are typically produced using traditional methods, which involve

mixing various medicinal herbs with rice flour and allowing the mixture to ferment under natural conditions. This process creates a favorable environment for the growth of diverse microorganisms, the most important of which are yeast species (e.g., *Saccharomyces cerevisiae*), molds (*Aspergillus*, *Rhizopus*, etc.), and lactic acid bacteria (including both lactic and acetic acid bacteria).

However, since the traditional production of starter cakes relies heavily on folk experience and lacks scientific and technical control, fermentation efficiency is often inconsistent, alcohol yield is low, and contamination risk is high. This underscores the urgent need for

*Corresponding author.

Email: hvqcanh@vnkgu.edu.vn

research and improvement in the production process.

Previous studies have demonstrated significant potential in isolating and applying indigenous microorganisms from starter cakes. Specifically, Ngo Thi Phuong Dung developed and applied defined starter cultures in the fermentation of black glutinous fermented glutinous rice, using *Amylomyces rouxii* and *Saccharomyces cerevisiae* as inoculum strains in the production of defined fermented glutinous rice starter.¹ Similarly, Nguyen Van Hue investigated the antioxidant and antibacterial properties of plant leaf extracts and applied them in the production of herbal starter cakes for fermented glutinous rice fermentation.² His findings showed that fermented glutinous rice produced with lab-made starter cakes containing leaf extract had alcohol concentrations comparable to wine fermented using traditional starter cakes obtained from local producers in Da Ban commune.

In addition, Doan Thi Kieu Tien and colleagues isolated *Saccharomyces cerevisiae* from Giac fruit wine (*Antidesma bunius*) and assessed its fermentation performance and storage conditions for Giac fruit wine.³ However, practical application studies of these strains under production conditions remain limited.

In the Mekong Delta region, which boasts a rich culinary culture and abundant agricultural resources, traditional starter cake villages are facing multiple challenges-from the loss of traditional knowledge to the pressure of competition from industrial products. In this context, conducting scientific research to evaluate and standardize starter cake production processes is a necessary direction. Such efforts not only help preserve and develop traditional crafts but also enhance product value, meeting the increasing demands for food safety and product quality.

From the issues outlined above, this study aims to investigate the production process of

fermented glutinous rice using isolated starter cultures, with key objectives including the application of selected microbial strains in a pilot fermentation process and evaluation of fermentation performance using local raw materials

2. MATERIALS AND METHOD

2.1. Materials and chemicals

Glutinous rice was procured from Vinh Tuy commune, Go Quao district, Kien Giang province.

Pure fermentation starters (banh men) were produced in the laboratory of Kien Giang University with the following microbial densities: molds at 10^7 CFU/g, yeasts at 10^7 CFU/g, and lactic acid bacteria at 10^8 CFU/g. Microorganisms present in the starter culture were isolated from traditional fermentation starters collected in Go Quao district, Kien Giang province. Molecular identification techniques revealed the presence of the yeast *Saccharomyces cerevisiae*, the filamentous fungus *Rhizopus oryzae*, and lactic acid bacterium *Lactiplantibacillus plantarum*. These strains were preserved and propagated in the Microbiology Laboratory of Kien Giang University.

Chemicals: 3,5-Dinitrosalicylic acid (China), NaOH 0.1N (Cemaco, Vietnam).

Culture media used for microbial propagation: YPD (Yeast extract Peptone Dextrose) medium, PDA (Potato Dextrose Agar) medium, MRS Broth medium, and various synthetic media from Himedia (India).

2.2. Methods

+ **Moisture (% w/w):** 10 g of fermented glutinous rice was determined for moisture content using a moisture balancer MOC-63u (Shimadzu, Japan).

+ **Brix:** 10 mL of water and fermented glutinous rice mixture, then measure Brix with Master-3T refractometer (Atago, Japan).

+ **The pH value:** Using 10 mL of water

and fermented glutinous rice mixture, dip the electrode of the pH meter MI150 MARTINI (Romania) into the solution to determine the pH.

+ **Alcohol content (% v/v):** Determine alcohol content by alcohol recovery distillation method, using alcohol distillation kit 2505200 (Witeg, Germany), then measure alcohol concentration CHG-3C33 (China) and convert to 20°C.⁴

+ **Titrateable acidity (as %lactic acid) (mg/mL):** Put 10 mL of water and fermented glutinous rice mixture into a test tube. Add 20 mL of distilled water. Add 1-2 drops of phenolphthalein. Shake well to mix the substances evenly. Titrate the solution with 0.1N NaOH until the solution turns pink, then stop.⁵

Lactic acid concentration is calculated in degrees Therner:

$$^{\circ}\text{T} = V_{\text{NaOH}} * 10$$

$$\% \text{ acid lactic (mg/mL)} = ^{\circ}\text{T} * 0.009$$

In which: $^{\circ}\text{T}$: degrees Therner, 1 $^{\circ}\text{T}$ corresponds to 9 mg of lactic acid. V_{NaOH} : volume of 0.1N NaOH consumed (mL)

Formula for calculating lactic acid concentration (mg/mL) = $V_{\text{NaOH}} * 10 * 0.009$

+ **Methanol content (%)** Using gas chromatography method according to TCVN 8010:2009

+ **Furfural content (%)** Using gas chromatography method according to TCVN 7886:2009

- The density of yeast, mold and lactic acid bacteria density by live count method on culture media:

The fermented glutinous rice samples were homogenized by placing 10 g of sample in a stomacher bag containing 90 mL of 0.85% NaCl solution, and the sample was ground for 1 min at 260 rpm. The samples were diluted to the appropriate concentration.

+ **Determining the density of yeast:** 0.1 mL of the diluted sample at a dilution of 10^{-5} was

spread on plates containing YPD agar and incubated in a UN110 incubator (Mettler, Germany) at 30 °C for about 48 h.⁶

+ **Determining the density of mold:** 0.1 mL of the diluted sample at a dilution of 10^{-5} was spread on plates containing PDA and incubated in a UN110 incubator (Mettler, Germany) at 37 °C for about 72 h.⁷

+ **Determining the density of lactic acid bacteria (LAB):** 0.1 mL of the diluted sample at a dilution of 10^{-5} was spread on plates containing MRS agar and incubated in a UN110 incubator (Mettler, Germany) at 37 °C for about 48 h.⁸

Formula for calculating density: $\text{CFU/g} = \frac{a \times n \times 10}{V}$ In which a: Number of colonies in 0.1 mL of sample; n: dilution.

+ **Sensory evaluation:** The numerical rating scale method was applied according to Vietnamese Standard TCVN 3215-79. The fermented glutinous rice samples were evaluated by a panel of 10 assessors, including lecturers and students from the Faculty of Food Science and Health. The evaluation was conducted using a 20-scores scale (0-20 scores) based on the following criteria: color, aroma, sweetness, mild sourness, and softness. Each attribute was equally weighted. The final result was calculated as the average score given by all panelists.

2.3. Data analysis

The results were processed and graphed using Microsoft Excel 2013 software (Microsoft Corporation, USA). The data were processed and statistically analyzed using Statgraphics Centurion XIX software (Statpoint Technologies Inc., USA).

3. RESULTS AND DISCUSSION

Sticky rice was finely ground and dried at 100°C for 24 hours to reduce moisture content and stabilize the raw material. The rice flour was then mixed with a microbial consortium consisting of mold, yeast, and lactic acid bacteria. These microorganisms were previously propagated in

suitable media: yeast in YPD, mold in PDA, and lactic acid bacteria in MRS broth, each incubated at 30°C for 24 hours.

After mixing the rice flour with the microbial cultures, sterilized water was added to achieve the desired moisture content. The mixture was thoroughly blended and formed into pellets. These pellets were incubated at 30°C for 24 hours to increase the microbial population. Following incubation, the fermented mass was dried at 42°C until the moisture content reached approximately 15%.

Traditional Fermented glutinous rice (Com ruou)
Production Process (for reference):

Sticky rice → First wash → Drained → First steaming (30 min) → Cooled → Second wash → Second steaming (30 min) → Cooled → Shaped into balls (hands moistened with salt water, salt/water ratio = 1:4) → Starter preparation → Starter ground finely → Evenly sprinkled over rice balls → Wrapped in banana leaves → Fermented in a plastic basket covered with PE film at room temperature → Collected fermented glutinous rice and extract. The final fermented glutinous rice product (com ruou) was

analyzed for various quality parameters, and the specific results are presented as follows:

3.1. Effect of the proportion of pure fermentation starters on the quality of fermented glutinous rice (Com ruou)

To evaluate the quality of com ruou using isolated fermentation starters at three different concentrations - 0.4%, 0.6%, and 0.8% (w/w based on cooked glutinous rice) - a traditional local production method was followed. The procedure included washing the rice, steaming twice (30 minutes each), cooling, shaping into balls with hands moistened in saltwater (ratio 1:4), sprinkling with finely ground fermentation starter, and fermenting the rice balls wrapped in banana leaves and placed in PE bags for 3 days to collect the sweet liquid extract. After fermentation, the samples were transported to the laboratory at Kien Giang University for quality analysis. The parameters assessed included pH, Brix value, alcohol content, mold, yeast and lactic acid bacteria counts, as well as sensory evaluation. The results aim to improve product quality and consistency for future commercialization. The findings are presented in Table 1.

Table 1. Effect of pure fermentation starters on the quality of fermented glutinous rice.

Concentration	Moisture (% w/w)	pH	Brix	Alcohol	Titratable acidity (mg/ml)
0.4%	47.5 ^b ± 1.8	3.98 ^b ± 0.06	40.33 ^b ± 0.57	10.2 ^b ± 0.4	0.75 ^a ± 0.05
0.6%	44.8 ^a ± 0.72	3.71 ^a ± 0.04	31.67 ^a ± 0.3	9 ^a ± 0.2	0.98 ^b ± 0.02
0.8%	43.76 ^a ± 0.76	3.91 ^b ± 0.08	32.67 ^a ± 0.5	8.8 ^a ± 0.2	0.89 ^b ± 0.1

Data are the mean values of triplicates. In the same column, different letters (a, b) indicate statistically significant differences at the 95% confidence level (statistical significance level $p < 0.05$).

Based on Table 1, the concentration of the isolated starter culture significantly influenced the moisture content of the fermented glutinous

rice. Increasing the starter culture concentration from 0.4% to 0.6% and 0.8% resulted in a corresponding decrease in moisture content

from 47.5% to 44.8% and 43.76%, respectively. The 0.4% sample exhibited the highest moisture content, which was statistically significantly different from the other two samples, while no significant difference was observed between the 0.6% and 0.8% samples. The moisture content of all samples ranged from 43% to 47% w/w. Moisture content reflects the water content in the food matrix, affecting quality, stability, and shelf life.⁹ Overall, the fermented glutinous rice in this study had moisture content below 50%, lower than that reported for Khao-Mak (50.76 - 53.04%).^{10,11} Table 1 also shows that the total soluble solids (TSS) of the finished fermented glutinous rice ranged from 31 to 40 °Brix. During fermentation, microorganisms secrete enzymes that metabolize starch into sugars, alcohol, and acids.^{12,13} In the initial phase, molds produce enzymes that degrade starch into soluble sugars. However, as fermentation progresses, the total soluble solids decrease due to the utilization of sugars as substrates by yeasts and other microorganisms.⁹ A similar decreasing trend has been observed during the production of Jiu Niang.¹⁴

Alcohol content of the fermented glutinous rice samples ranged from 8% to 10% v/v, with statistically significant differences observed among them. Saccharification by molds and alcoholic fermentation by yeasts convert glucose into alcohol.⁷ A small amount of alcohol can also be produced by lactic acid bacteria during fermentation at temperatures above 30 °C in the presence of maltose.¹⁵ Compared to previous studies, alcohol levels reported here are relatively high. For example, Khao-Mak from Thailand reached only 2.15 - 2.58% v/v after 3 days of fermentation,¹⁰ while other studies reported alcohol concentrations below 0.5% v/v at day 3, gradually increasing to 1 - 2% v/v by day 7.¹⁶ This discrepancy may be attributed to differences in raw materials.¹⁶

The lactic acid content in the fermented glutinous rice samples ranged from 0.75 to 0.98 mg/mL. An inverse correlation was observed

between alcohol and lactic acid concentrations: alcohol accumulation inhibits acid production, thereby helping to control acidity and reduce spoilage.¹⁵ The 0.4% starter culture sample exhibited the lowest lactic acid concentration (0.75 mg/mL), whereas the 0.6% sample had the highest (0.98 mg/mL), similar to the lactic acid content reported in Khao-Mak fermentation after 3 days.² Jiu Niang also contains various organic acids, primarily lactic acid, along with malic, fumaric, acetic, tartaric, citric, and succinic acids, with total concentrations increasing over time and slightly decreasing after 60 hours.¹⁴ This phenomenon is related to the complex activity of the microbial community in the starter culture or possible contamination.¹⁷

pH values of fermented glutinous rice samples in this study ranged from 3.71 to 3.98, reflecting the level of organic acid accumulation during fermentation. There was an inverse relationship between pH and titratable acidity, as lactic acid, produced by lactic acid bacteria, accumulated, the pH decreased. Although the pH variations were relatively small, even slight changes could significantly affect the sensory attributes and stability of the final product. This reflects the relationship between pH and the ionization strength of acids. In the case of Khao-Mak made from black rice, the pH decreased from 6.00 to 4.00–4.27 after 3 days of fermentation.¹⁸ Yeasts and molds participate in the conversion of sugars into alcohol and acids, contributing to the pH reduction and creating an antimicrobial environment that ensures food safety.¹⁷ Overall, the quality of fermented glutinous rice varies depending on the source of the starter culture, traditional processing methods, environmental conditions, and fermentation duration.

In addition to physicochemical parameters such as moisture content, pH, acid concentration, Brix value, and alcohol content, the fermented glutinous rice product was also subjected to sensory evaluation to assess consumer preference. The results of the sensory evaluation are presented in Table 2.

Table 2. Effect of pure fermentation starters concentration on the sensory quality of fermented glutinous rice product.

Concentration	Average Sensory Scores	Methanol (%)	Furfurol (%)
0.4%	18.41 ^b ± 0.27	ND	ND
0.6%	16.41 ^a ± 0.34	ND	ND
0.8%	16.0 ^a ± 0.25	ND	ND

Note: “ND: no detected”; *Data are the mean values of triplicates. In the same column, different letters (a, b) indicate statistically significant differences at the 95% confidence level (statistical significance level $p < 0.05$).*

The average sensory score at a starter culture concentration of 0.4% was 18.41 ± 0.27 , which was statistically significantly different from those at 0.6% (16.41 ± 0.34) and 0.8% (16.00 ± 0.25). No significant difference was observed between the sensory scores of the 0.6% and 0.8% samples. These results indicate that the product achieved the highest sensory acceptance at a starter culture concentration of 0.4%.

Moreover, the analysis showed that two undesirable compounds, methanol and furfural, were not detected in the fermented glutinous rice product. These compounds are known to

cause headaches and have neurotoxic effects on consumers. Therefore, the absence of these substances in the experimental samples is highly significant for future commercialization.

In addition to physicochemical and sensory parameters, microbiological characteristics of the fermented glutinous rice samples were also analyzed, including determining the density of molds, yeasts, and lactic acid bacteria. This provided an overview of the microbial populations present in the product. The results of microbial enumeration are presented in Table 3.

Table 3. Effect of pure fermentation starters concentration on microbial population in the product.

Concentration	Molds (Log ₁₀ CFU/g)	Yeasts (Log ₁₀ CFU/g)	Lactic acid bacteria (Log ₁₀ CFU/g)
0.4%	7.27 ^c ± 0.02	7.28 ^c ± 0.01	8.22 ^a ± 0.01
0.6%	6.22 ^a ± 0.02	7.27 ^b ± 0.01	8.27 ^b ± 0.02
0.8%	6.92 ^b ± 0.01	7.20 ^a ± 0.01	8.24 ^a ± 0.01

Data are the mean values of triplicates. In the same column, different letters (a, b) indicate statistically significant differences at the 95% confidence level (statistical significance level $p < 0.05$).

The analysis results presented in Table 3 indicate that the fermented glutinous rice sample with 0.4% isolated starter culture had the highest yeast and mold counts, at $7.27 \log_{10}$ CFU/g and $7.28 \log_{10}$ CFU/g, respectively, while the lowest counts were observed in the 0.6% sample, with yeast and mold counts of $6.22 \log_{10}$ CFU/g and $7.27 \log_{10}$ CFU/g, respectively. These findings

confirm the presence of both molds and yeasts in the fermented glutinous rice samples. These two groups of microorganisms play key roles and exhibit principal activities during saccharification and alcoholic fermentation processes.^{19,20}

Similarly, a study on Khao-Mak fermentation⁶ reported comparable results, where at the end of the fermentation period (day 3),

total mold and yeast counts were 7.00×10^2 CFU/g and 5.40×10^5 CFU/g, respectively.¹¹ Additionally, lactic acid bacteria were also detected. Besides molds, certain bacteria are capable of producing enzymes that hydrolyze starch in raw materials into sugars, which yeasts can subsequently utilize for alcohol production.²¹

For this reason, research on the production of Tapai-a traditional Indonesian product similar to Vietnamese fermented glutinous rice has explored supplementation with *Lactobacillus plantarum* strains alongside starter cultures. The results demonstrated that Tapai fermented

with *L. plantarum* exhibited slightly different characteristics compared to traditional Tapai and received higher acceptance scores from sensory panelists.²²

3.2. Effect of fermentation time on the quality of fermented glutinous rice

The fermentation process of fermented glutinous rice exhibited significant changes in physicochemical parameters over time. Table 4 presents data on Brix value, pH, alcohol content, and sensory scores at fermentation time of 2, 3, and 4 days.

Table 4. Effect of fermentation time on the quality of fermented glutinous rice.

Parameter	Fermentation Time (days)		
	2	3	4
Brix (°Brix)	39.4 ^c ± 0.52	38.1 ^b ± 0.26	35.6 ^a ± 0.2
pH	3.82 ^a ± 0.02	4.05 ^c ± 0.04	3.91 ^b ± 0.01
Alcohol (% v/v)	8.0 ^a ± 0.2	10.67 ^b ± 0.58	11.02 ^b ± 0.57
Sensory Evaluation (Scores)	14.50 ^a ± 0.32	17.44 ^c ± 0.14	16.25 ^b ± 0.26
Moisture (% w/w)	44.3 ^a ± 0.67	47.5 ^b ± 0.05	49.2 ^c ± 0.22
Titrateable acidity (mg/mL)	0.87 ^a ± 0.05	0.99 ^{ab} ± 0.01	1.05 ^b ± 0.05
Methanol (%)	ND	ND	ND
Furfurol (%)	ND	ND	ND

Note: “ND: no detected”; Data are the mean values of triplicates. In the same row, different letters (a, b) indicate statistically significant differences at the 95% confidence level (statistical significance level $p < 0.05$).

The study results demonstrated that fermentation time significantly influenced the physicochemical and sensory parameters of the product. The Brix value gradually decreased from 39.4 °Brix on day 2 to 38.1 °Brix on day 3 and further to 35.6 °Brix on day 4. This reduction was statistically significant and reflects the conversion of sugars into alcohol and other by-products. The pH of the samples showed fluctuations, with a slight increase from 3.82 to 4.05 on day 3, followed by a decrease to 3.91 on day 4. These changes may be associated with the formation and degradation of organic acids during fermentation, although the pH remained within the weak acid range appropriate for fermented food products.

Alcohol content in the product increased from 8.0% on day 2 to 10.67% on day 3, reaching 11.02% on day 4. The difference between day 2

and the subsequent time points was statistically significant; however, the variation between days 3 and 4 was not significant, indicating that the rate of alcohol biosynthesis peaked around day 3. The sensory score rose sharply from 14.50 on day 2 to 17.44 on day 3, followed by a slight decrease to 16.25 on day 4. The sample fermented for 3 days was rated as having the best sensory quality, reflecting a balanced combination of flavor, alcohol content, and acidity.

The moisture content of the product gradually increased over time, from 44.3% to 49.2%, reflecting the breakdown and dissolution of compounds during raw material hydrolysis. Concurrently, the total acid content also showed a slight increase from 0.87 to 1.05 mg/mL, consistent with the accumulation of organic acids due to microbial activity. Notably, neither

malcohol nor furfural was detected in any of the samples, confirming the safety of the fermentation process.

In addition to physicochemical and sensory parameters, microbiological indicators of the fermented glutinous rice samples were also

analyzed, including the enumeration of molds, yeasts, and lactic acid bacteria. This provided an overall overview of the microbial populations present in the fermented glutinous rice product. The results of the microbial counts are presented in Table 5.

Table 5. Effect of fermentation time on microbial population in the product.

Fermentation Time (days)	Molds (Log ₁₀ CFU/g)	Yeasts (Log ₁₀ CFU/g)	Lactic acid bacteria (Log ₁₀ CFU/g)
2	7.01 ^c ± 0.05	7.03 ^c ± 0.01	8.14 ^a ± 0.01
3	6.13 ^b ± 0.03	7.15 ^b ± 0.01	8.23 ^b ± 0.02
4	5.92 ^a ± 0.02	7.6 ^a ± 0.01	8.34 ^c ± 0.01

Data are the mean values of triplicates. In the same row, different letters (a, b) indicate statistically significant differences at the 95% confidence level (statistical significance level $p < 0.05$).

During the fermentation process, significant changes in the population densities of microbial groups were observed over time. The mold count gradually decreased from 7.01 log₁₀ CFU/g on day 2 to 6.13 log₁₀ CFU/g on day 3, reaching the lowest level of 5.92 log₁₀ CFU/g on day 4. This reduction was statistically significant, as indicated by different letter notations on the mean values ($p < 0.05$). In contrast, yeast populations increased significantly over time, from 7.03 log₁₀ CFU/g on day 2 to 7.15 log₁₀ CFU/g on day 3 and 7.60 log₁₀ CFU/g on day 4, with the increase also showing statistical significance. Similarly, lactic acid bacteria exhibited strong growth trends, with counts rising from 8.14 log₁₀ CFU/g to 8.23 and 8.34 log₁₀ CFU/g on days 2, 3, and 4, respectively. Mean values with different letter notations confirmed significant differences between days ($p < 0.05$).

These results indicate that the fermentation process created favorable conditions for the proliferation of yeasts and lactic acid bacteria-two beneficial microbial groups-while substantially inhibiting mold growth, thereby contributing to the quality and microbiological

safety of the product. This trend aligns with international reports on alcoholic fermentation. According to Fleet,²³ during wine fermentation, *Saccharomyces cerevisiae* is the dominant yeast species, which grows vigorously and produces a harsh environment (alcohol and low pH) that suppresses undesirable microorganisms such as molds and aerobic bacteria. Yoon similarly reported approximately 4.6×10^7 CFU/mL of *Saccharomyces cerevisiae* present in fermented glutinous rice water after 48 hours of fermentation.²⁴

Collectively, these findings suggest that a fermentation period of 3 days is optimal, yielding high alcohol content, favorable sensory qualities, and satisfactory safety parameters. This duration is recommended for practical application to maximize product quality and food safety.

Thus, the optimal fermentation time for fermented glutinous rice is 3 days using a 0.4% starter culture concentration, ensuring ideal quality parameters such as pH, Brix, alcohol concentration, acid content, and counts of molds, yeasts, and lactic acid bacteria. Additionally, the final product received positive sensory evaluations from consumers.

4. CONCLUSIONS

The study applied pure microbial strains isolated from traditional fermentation starters collected in Go Quao district, Kien Giang province, for the production of fermented glutinous rice. The results indicated that the ratio of fermentation starter used and fermentation time were two critical factors affecting the sensory, physicochemical, and microbiological quality of the final product. Among the tested concentrations of pure fermentation starter (0.4%, 0.6%, and 0.8%), the 0.4% level yielded optimal results in terms of sensory quality and fermentation performance, with a high ethanol content (10.2% v/v), appropriate moisture and pH values, balanced lactic acid levels, and no detection of harmful compounds such as methanol or furfural. Additionally, samples at this concentration showed high yeast and mold counts, indicating effective fermentation.

Regarding fermentation time, three durations (2 - 4 days) were evaluated, with 3 days determined to be optimal, showing a balance of physicochemical parameters (pH, Brix, moisture, ethanol, acid), favorable sensory characteristics, and viable counts of beneficial microorganisms. Notably, the simultaneous increase in yeast and lactic acid bacteria populations and the decline in mold counts reflected the establishment of a stable microbial community, suitable for the production of safe fermented products.

The findings provide a scientific basis for standardizing the use of pure starter cultures in terms of ratio and fermentation time, while enhancing the stability and sensory quality of traditional fermented glutinous rice. More importantly, this research contributes to the preservation and sustainable development of traditional starter-making practices-a valuable cultural heritage of the Mekong Delta-through a scientific, safe, and commercially viable approach.

REFERENCES

1. N. T. P. Dung, F. Rombouts, M. Nout. Characteristics of some traditional Vietnamese starch-based rice wine fermentation starters (men), *LWT-Food Science and Technology*, **2007**, *40*, 130-135.
2. N. V. Hue, N. T. V. Anh, and H. T. Kim. Antioxidant and antibacterial activities of leaf extracts and their application in starter cake production for leaf yeast wine fermentation (Vietnamese), *Hue University Journal of Science: Agriculture and Rural Development*, **2021**, *130*, 57-73.
3. D. T. K. Tien, H. T. N. Mi, H. X. Phong, N. N. Thanh, B. H. D. Long, H. T. Toan, N. T. P. Dung. Isolation, selection, and application of thermotolerant yeast in *Giac* fruit wine fermentation (Vietnamese), *Can Tho University Journal of Science*, **2021**, *57*, 132-143.
4. N. D. Thuong, N. T. Hang. *Technology of alcohol production and analysis (Vietnamese)*, Science and Technics Publishing House, Ha Noi, 2007.
5. N. T. M. Hang, N. M. Thu. Isolation and selection of some lactic acid bacteria strains capable of synthesizing amylase and bacteriocin (Vietnamese), *Journal of Forestry Science and Technology*, **2013**, 003-010.
6. C. P. Kurtzman, J. W. Fell, T. Boekhout, V. Robert. Methods for isolation, phenotypic characterization and maintenance of yeasts. *The yeasts (fifth edition)*, Elsevier, USA, 2011.
7. L. V. X. Cong, Z. Q. Huang, W. Zhang, P. F. Rao, L. Ni. Identification and characterization of filamentous fungi isolated from fermentation starters for *Hong Qu* glutinous rice wine brewing, *The Journal of General and Applied Microbiology*, **2012**, *58*, 33-42.
8. T. T. X. Mai. *Molecular biology laboratory manual (Vietnamese)*, Can Tho University Publishing House, Can Tho, 2010.

9. H. T. Thanh, H. Q. Huy, N. T. N. Anh, L. M. Chau, N. N. Thanh, B. H. D. Long, H. X. Phong. Investigation of quality and isolation of yeast and mold from fermented glutinous rice (com ruou) produced in Can Tho City (Vietnamese), *Can Tho University Journal of Science*, **2024**, *60*, 461-472.
10. V. Surojanametakul, W. Panthavee, P. Satmalee, N. Phomkaivon, T. Yoshihashi. Effect of traditional dried starter culture on morphological, chemical, and physicochemical properties of sweet fermented glutinous rice products, *Journal of Agricultural Science*, **2019**, *11*(6), 43-51.
11. V. Surojanametakul, W. Panthavee, P. Satmalee, N. Phomkaivon, T. Yoshihashi. Effect of traditional dried starter culture on morphological, chemical, and physicochemical properties of sweet fermented glutinous rice products, *Journal of Agricultural Science*, **2024**, *11*, 1-43.
12. N. D. Luong. *Microbial technology, volume 3: traditional fermented foods (Vietnamese)*, Ho Chi Minh City University of Technology Publishing House, Ho Chi Minh, 2005.
13. L. D. Pham. *Industrial Yeasts (Vietnamese)*, Science and Technology Publishing House, Hanoi, 2006.
14. Y. Xu, Y. Jin, J. Su, N. Yang, X. Xu, Z. Jin, B. Cui, F. Wu. Changes in the nutritional value, flavor, and antioxidant activity of brown glutinous rice during fermentation, *Food Bioscience*, **2021**, *43*, 101273.
15. C. Åkerberg, K. Hofvendahl, G. Zacchi, B. H. Hågerdal. Modelling the influence of pH, temperature, glucose, and lactic acid concentrations on the kinetics of lactic acid production by *Lactococcus lactis* ssp. *lactis* ATCC 19435 in whole-wheat flour, *Applied Microbiology and Biotechnology*, **1998**, *49*, 682-690.
16. J. Wongsas, V. Rungsardthong, T. Yasutomo. *Production and analysis of volatile flavor compounds in sweet fermented rice (Khao Mak)*, The 4th International Conference on Engineering, Applied Sciences and Technology (ICEAST 2018), Phuket, Thailand, 2018.
17. C. Zhao, W. Su, Y. Mu, L. Jiang, Y. Mu. Correlations between microbiota with physicochemical properties and volatile flavor components in black glutinous rice wine fermentation, *Food Research International*, **2020**, *138*, 109800.
18. A. Manosroi, W. Ruksiriwanich, B. O. Kietthanakorn, W. Manosroi, J. Manosroi. Relationship between biological activities and bioactive compounds in fermented rice sap, *Food Research International*, **2011**, *44*, 2757-2765.
19. C. Hesseltine, R. Rogers, F. Winarno. Microbiological studies on amylolytic oriental fermentation starters, *Mycopathologia*, **1998**, *101*, 141-155.
20. C. Nwosu, P. Ojmelukwe. Improvement of the traditional method of *ogiri* production and identification of the micro-organisms associated with the fermentation process, *Plant Foods for Human Nutrition*, **1993**, *43*, 267-272.
21. L. Hong, G. Fu, T. Liu, Y. Chen, S. Wu, W. Cai, Z. Xie, Y. Wan. Functional microbial agents enhance ethanol contents and regulate the volatile compounds in Chinese *Baijiu*, *Food Bioscience*, **2021**, *44*, 101411.
22. Y. Yusmarini, V. S. Johan, S. Fitriani, R. Rahmayuni, V. F. Artanti, U. Pato. Characteristics of probiotic *tapai* made by the addition of *Lactobacillus plantarum* 1, *International Journal of Agricultural Technology*, **2019**, *15*(1), 195-206.
23. G. H. Fleet. Yeast interactions and wine flavour, *International Journal of Food Microbiology*, **2003**, *86*(1-2), 11-22.
24. S. S. Yoon, J. A. Choi, K. H. Kim, T. S. Song, Y. S. Park. Populations and potential association of *Saccharomyces cerevisiae* with lactic acid bacteria in naturally fermented Korean rice wine, *Food Science and Biotechnology*, **2012**, *21*, 419-424.



Một đặc trưng của không gian kiểu Zygmund và áp dụng

Thái Thuần Quang^{1,*} Nguyễn Văn Đại²

¹Khoa Toán và Thống kê, Trường Đại học Quy Nhơn, Việt Nam

²Khoa Sư phạm, Trường Đại học Quy Nhơn, Việt Nam

Ngày nhận bài: 26/06/2025; Ngày sửa bài: 12/08/2025

Ngày nhận đăng: 19/08/2025; Ngày xuất bản: 28/10/2025

TÓM TẮT

Trong bài báo này, chúng tôi nghiên cứu một số điều kiện để không gian kiểu Zygmund Z_ω , với ω là một trọng chuẩn tắc trên hình cầu đơn vị $\mathbb{B}$ trong $\mathbb{C}^n$, trở thành một không gian nhỏ, ổn định biên và bất biến dưới các tự đẳng cấu. Chúng tôi áp dụng kết quả này để phân tích mối quan hệ giữa tính bị chặn và tính compact của các toán tử hợp liên tục $W_{\psi,\varphi}$, từ $\mathcal{B}_\omega$ vào Z_ω và trên Z_ω .

Từ khóa: Không gian Bloch, không gian Zygmund, toán tử hợp có trọng, tính bị chặn, tính compact.

*Tác giả liên hệ chính.

Email: thaithuanquang@qnu.edu.vn

A characterization of Zygmund-type spaces and its application

Thai Thuan Quang^{1,*} Nguyen Van Dai²

¹Department of Mathematics and Statistics, Quy Nhon University, Vietnam

²Department of Education, Quy Nhon University, Vietnam

Received: 26/06/2025; Revised: 12/08/2025

Accepted: 19/08/2025; Published: 28/10/2025

ABSTRACT

In this paper, we examine the conditions under which a Zygmund-type space $\mathcal{Z}_\omega$, where ω is a normal weight on the unit ball $\mathbb{B}$ of $\mathbb{C}^n$, becomes a small space that is boundary regular and invariant under automorphisms. These results are then applied to analyze the relationship between the boundedness and compactness of weighted composition operators $W_{\psi,\varphi}$, defined by $f \mapsto \psi \cdot (f \circ \varphi)$, acting from the Bloch-type space $\mathcal{B}_\omega$ to the Zygmund-type space $\mathcal{Z}_\omega$, as well as from $\mathcal{Z}_\omega$ into itself.

Keywords: Bloch spaces, Zygmund spaces, weighted composition operators, boundedness, compactness

1. INTRODUCTION

Given a natural number n , let us consider the open unit ball $\mathbb{B}$ in $\mathbb{C}^n$ and $H(\mathbb{B})$ the space of all holomorphic functions in $\mathbb{B}$. The vectors $e_1 = (1, 0, \dots, 0)$, $e_2 = (0, 1, 0, \dots, 0), \dots$, $e_n = (0, \dots, 0, 1)$ form the standard basis for $\mathbb{C}^n$.

Throughout this paper, for any $z = (z_1, \dots, z_n)$ and $w = (w_1, \dots, w_n)$ in $\mathbb{C}^n$, we use $\langle z, w \rangle = \sum_{k=1}^n z_k \bar{w}_k$, to denote their standard inner product, and write $|z| = \sqrt{\langle z, z \rangle} = \sqrt{|z_1|^2 + \dots + |z_n|^2}$ for the corresponding Euclidean norm.

For $f \in H(\mathbb{B})$, let

$$\begin{aligned} \nabla_z f(z) &= \left(\frac{\partial f}{\partial z_1}(z), \dots, \frac{\partial f}{\partial z_n}(z) \right), \\ Rf(z) &= \langle \nabla f(z), \bar{z} \rangle, \quad z \in \mathbb{B}. \end{aligned}$$

Let $\mathbb{D}$ denote the unit disk of $\mathbb{C}$. If $f \in H(\mathbb{D})$ satisfies $\sup_{z \in \mathbb{D}} (1 - |z|^2) |f''(z)| < \infty$ then f is said to belong to the Zygmund space. In this definition, $1 - |z|^2$ acts as a weight function, which was later generalized to $(1 - |z|^2)^\alpha$ for all $\alpha > 0$.

A positive continuous function ω defined on the interval $[0, 1)$ is said to be normal if there exist constants $0 \leq \delta < 1$ and $0 < a < b < \infty$ such that

$$\frac{\omega(t)}{(1-t)^a} \text{ is decreasing on } [\delta, 1), \quad (W_1)$$

$$\lim_{t \rightarrow 1} \frac{\omega(t)}{(1-t)^a} = 0,$$

$$\frac{\omega(t)}{(1-t)^b} \text{ is increasing on } [\delta, 1), \quad (W_2)$$

$$\lim_{t \rightarrow 1} \frac{\omega(t)}{(1-t)^b} = \infty.$$

*Corresponding author.

Email: thaithuanquang@qnu.edu.vn

If we say that a function $\omega : \mathbb{B} \rightarrow [0, \infty)$ is normal, we also assume that it is radial, that is, $\omega(z) = \omega(|z|)$ for every $z \in \mathbb{B}$. Strictly positive continuous functions on $\mathbb{B}$ are referred to as weights.

We define Bloch-type space $\mathcal{B}_\omega$, Zygmund-type space $\mathcal{Z}_\omega$, respectively, as follows:

$$\mathcal{B}_\omega = \left\{ f \in H(\mathbb{B}) : \|f\|_{s\mathcal{B}_\omega} < \infty \right\},$$

$$\mathcal{Z}_\omega = \left\{ f \in H(\mathbb{B}) : \|f\|_{s\mathcal{Z}_\omega} < \infty \right\},$$

where

$$\|f\|_{s\mathcal{B}_\omega} = \sup_{z \in \mathbb{B}_n} \omega(z) |(Rf)(z)|,$$

$$\|f\|_{s\mathcal{Z}_\omega} = \sup_{z \in \mathbb{B}_n} \omega(z) |\nabla(Rf)(z)|$$

are seminorms on $\mathcal{B}_\omega$ and $\mathcal{Z}_\omega$, respectively. The spaces $\mathcal{B}_\omega$, $\mathcal{Z}_\omega$ are endowed with Banach space structures via the norm

$$\|f\|_{\mathcal{B}_\omega} = |f(0)| + \|f\|_{s\mathcal{B}_\omega},$$

$$\|f\|_{\mathcal{Z}_\omega} = |f(0)| + \|f\|_{s\mathcal{Z}_\omega}.$$

The space $\mathcal{Z}_\omega$ generalizes the classical Zygmund space, which was introduced in the paper.¹

Define $S(\mathbb{B})$ as the set of holomorphic self-maps of $\mathbb{B}$. Given $\psi \in H(\mathbb{B})$ and $\varphi \in S(\mathbb{B})$, the weighted composition operator $W_{\psi,\varphi} : E \rightarrow F$ is defined by

$$W_{\psi,\varphi}(f) := \psi \cdot (f \circ \varphi), \quad \text{for } f \in E,$$

where E and F are Banach spaces consisting of holomorphic functions on $\mathbb{B}$. It may be regarded as a generalization of multiplication and composition operators.

The theory of composition operators, both weighted and unweighted, has its origins in the previous century. The boundedness, compactness, essential norm, and spectral properties are always the highlights of research of composition operators. Book² is a good reference for studying the composition operators on classical spaces of analytic functions. Furthermore, the theory relies on the

theory of analytic functions on the unit disk, which provides a convenient foundation.

Composition operators mapping into the classical Zygmund were studied in the papers.^{3–9} Many scholars have discussed similar problems (see in the papers,^{10–17} etc.)

However, for abstract normal weight especially in high dimensions, when investigating and using the properties (for example, discussing weighted/unweighted composition operator of the Zygmund type space, we often encounter some obstacles. This partly explains why the boundedness and compactness criteria for $W_{\psi,\varphi}$ between Zygmund-type spaces (normal weight cases) have not been extensively investigated to date. In order to overcome these obstacles, we need a variety of means or techniques.

Motivated by the above-mentioned discussions and the previous investigations, the purpose of this paper is to uncover additional characteristics of Zygmund-type spaces and serve them as technical tools to solve the problem of the relationship between the boundedness, compactness of weighted composition operators from a Bloch-type space $\mathcal{B}_\omega$ into the Zygmund-type space $\mathcal{Z}_\omega$ and from $\mathcal{Z}_\omega$ into itself.

Section 2 provides a sufficient condition on the normal weight ω ensuring that $\mathcal{Z}_\omega$ is an automorphism invariant boundary regular small space. A key motivation for this section comes from a result of Shapiro¹⁸ (and Theorem 4.5 in the paper²), which asserts that $\|\varphi\|_\infty < 1$ is a necessary condition for C_φ to be compact on any “suitably small” Banach space. According to Shapiro,¹⁸ four axioms must be satisfied for a space to qualify as appropriately small. Among these, two axioms are fundamental, concerning norm naturality and space nontriviality, while the other two regulate the size of the spaces. Specifically, the boundary regularity axiom makes the spaces small by ensuring continuous boundary extension, and

the automorphism-invariance axiom prevents them from being excessively small. For further details, refer to paper¹⁸ or paper.²

Building on the results obtained in the previous section, Section 3 establishes the connections between the boundedness and compactness of weighted composition operators acting from $\mathcal{B}_\omega$ to $\mathcal{Z}_\omega$, and those acting on $\mathcal{Z}_\omega$ itself.

In this paper, we use the notation $a \lesssim b$ to denote that $a \leq Cb$, and $a \asymp b$ to indicate that $C^{-1}b \leq a \leq Cb$, where $C > 0$ is an inessential constant, with all quantities a and b assumed to be non-negative.

2. A CHARACTERIZATION OF ZYGMUND-TYPE SPACES

This section is devoted to the study of the properties “small” and “automorphism invariant boundary regular” of the Zygmund-type spaces which will be necessary in establishing one of our main result.

For a normal weight ω on $\mathbb{B}$ we use there certain quantities, which will be used in this work:

$$I_\omega^1(z) := \int_0^{|z|} \frac{dt}{\omega(t)},$$

$$I_\omega^2(z) := \int_0^{|z|} \left(\int_0^t \frac{ds}{\omega(s)} \right) dt, \quad z \in \overline{\mathbb{B}}.$$

Remark 2.1. Since ω is positive and continuous, it follows that $m_{\omega,\delta} := \min_{t \in [0,\delta]} \omega(t) > 0$. In addition, by (W_1) , ω is strictly decreasing on $[\delta, 1)$, so $\max_{t \in [0,1)} \omega(t) =: M_\omega < \infty$. Consequently, one can easily verify that

$$\omega(z)I_\omega^1(z) < R_\omega := \delta \frac{M_\omega}{m_{\omega,\delta}} + 1 - \delta < \infty \quad (2.1)$$

and, hence,

$$\omega(z)I_\omega^2(z) < |z|R_\omega < R_\omega < \infty \quad (2.2)$$

for every $z \in \mathbb{B} \setminus \{0\}$.

Proposition 2.1 ⁽¹⁷⁾. For every normal weight ω on $\mathbb{B}$ we have

$$\begin{aligned} \mathcal{Z}_\omega &= \mathcal{Z}_\omega^R := \left\{ f \in H(\mathbb{B}) : \|f\|_{\mathcal{Z}_\omega^R} < \infty \right\} \\ &= \mathcal{Z}_\omega^\nabla := \left\{ f \in H(\mathbb{B}) : \|f\|_{\mathcal{Z}_\omega^\nabla} < \infty \right\} \end{aligned}$$

and $\|\cdot\|_{\mathcal{Z}_\omega} \cong \|\cdot\|_{\mathcal{Z}_\omega^R} \cong \|\cdot\|_{\mathcal{Z}_\omega^\nabla}$, where

$$\begin{aligned} R^{(2)}f &= R(Rf), \\ |\nabla^{(2)}f(z)| &= \left(\sum_{i,j=1}^n \left| \frac{\partial^2 f}{\partial z_i \partial z_j}(z) \right|^2 \right)^{\frac{1}{2}}, \\ \|f\|_{\mathcal{Z}_\omega^R} &:= |f(0)| + \sup_{z \in \mathbb{B}_n} \omega(z) |R^{(2)}f(z)|, \\ \|f\|_{\mathcal{Z}_\omega^\nabla} &:= |f(0)| + \sup_{z \in \mathbb{B}_n} \omega(z) |\nabla^{(2)}f(z)|, \end{aligned}$$

for every $f \in \mathcal{Z}_\omega$.

In this paper, let us write simply we denote $\mathcal{Z}_\omega$ for the complex $(\mathcal{Z}_\omega, \|\cdot\|_{\mathcal{Z}_\omega^R})$.

Lemma 2.2. Let ω be a normal weight on $\mathbb{B}$. Then there exists $C > 0$ such that for every $f \in \mathcal{Z}_\omega$ and every $z \in \mathbb{B}$,

$$\begin{aligned} |Rf(z)| &\leq CI_\omega^1(z) \|f\|_{\mathcal{Z}_\omega}, \\ |\nabla f(z)| &\leq C(1 + I_\omega^1(z)) \|f\|_{\mathcal{Z}_\omega}; \end{aligned} \quad (2.3)$$

and

$$|f(z)| \leq |f(0)| + CI_\omega^2(z) \|f\|_{\mathcal{Z}_\omega}. \quad (2.4)$$

Proof. The estimate (2.3) follows from¹⁹ which says there is $C > 0$ such that for every $f \in \mathcal{B}_\omega$ and for all $z \in \mathbb{B}$,

$$|f(z)| \leq C(1 + I_\omega^1(z)) \|f\|_{\mathcal{B}_\omega}. \quad (2.5)$$

Then by (2.3) and (2.5) again we obtain (2.4). $\square$

Note that, in fact, by using (2.5) the estimate for $|\nabla f(z)|$ in (2.3) can be replaced by

$$|\nabla f(z)| \lesssim (1 + I_\omega^1(z)) |\nabla f(0)| + I_\omega^1(z) \|f\|_{\mathcal{Z}_\omega}. \quad (2.6)$$

Now, by $\text{Aut}(\mathbb{B})$, we denote the automorphism group of $\mathbb{B}$ that consists of all biholomorphic mappings of $\mathbb{B}$. It is well known

that a mapping $\varphi \in \text{Aut}(\mathbb{B})$ is a unitary transformation of $\mathbb{C}^n$ if and only if $\varphi(0) = 0$ (see ²⁰). For any $\alpha \in \mathbb{B} \setminus \{0\}$, we define

$$\varphi_\alpha(z) = \frac{\alpha - P_\alpha(z) - s_\alpha Q_\alpha(z)}{1 - \langle z, \alpha \rangle}, \quad z \in \mathbb{B}, \quad (2.7)$$

where $s_\alpha = \sqrt{1 - |\alpha|^2}$, $P_\alpha(z) = \frac{\langle z, \alpha \rangle}{|\alpha|^2} \alpha$ and $Q_\alpha(z) = z - \frac{\langle z, \alpha \rangle}{|\alpha|^2} \alpha$ for all $z \in \mathbb{B}$.

For $\alpha = 0$, we simply set $\varphi_\alpha(z) = -z$. Clearly, each φ_α is holomorphic from $\mathbb{B}$ into $\mathbb{C}^n$. It is also well known that each φ_α is a homeomorphism of $\overline{\mathbb{B}}$ onto itself, and every automorphism φ of $\mathbb{B}$ can be represented as $\varphi = \varphi_\alpha U$, with U a unitary transformation on $\mathbb{C}^n$.

Theorem 2.3. Let ω be normal weight on $\mathbb{B}$ such that $I_\omega^2(1) < \infty$. Then the space $\mathcal{Z}_\omega$ is an automorphism invariant boundary regular small space in the following sense:

- (i) All functions in $\mathcal{Z}_\omega$ can be continuously extended to $\overline{\mathbb{B}}$,
- (ii) All polynomials are contained in $\mathcal{Z}_\omega$,
- (iii) Evaluation at every point in $\mathbb{B}$ defines a bounded linear functional,
- (iv) For any $\varphi \in \text{Aut}(\mathbb{B})$ and $f \in \mathcal{Z}_\omega$, we have $f \circ \varphi \in \mathcal{Z}_\omega$.

Remark 2.2. Axioms (i) and (iii) ensure that convergence in the $\mathcal{Z}_\omega$ norm implies convergence in the sup norm; that is, the identity map from $(\mathcal{Z}_\omega, \|\cdot\|_{\mathcal{Z}_\omega})$ to $(\mathcal{Z}_\omega, \|\cdot\|_\infty)$ is continuous by the closed graph theorem. Furthermore, using the closed graph theorem along with axiom (iii), one can show that axiom (iv) ensures that C_φ is bounded on $\mathcal{Z}_\omega$ for any conformal automorphism φ of $\mathbb{B}$.

Proof. It is straightforward from the definitions to verify that (ii) and (iii) hold for $\mathcal{Z}_\omega$. Under the condition $I_\omega^1(1) < \infty$ the space $\mathcal{Z}_\omega$ satisfies (i) (see paper ¹⁵).

To verify that (iv) holds, we need to show that for any conformal automorphism

$\varphi = \varphi_a U = (\varphi_1, \dots, \varphi_n)$ of $\mathbb{B}$, if $f \in \mathcal{Z}_\omega$, then $f \circ \varphi \in \mathcal{Z}_\omega$, where a is a point in $\mathbb{B}$ and U is a unitary transformation of $\mathbb{C}^n$. Without loss of generality, we can assume that $\varphi = \varphi_a$ for some $a \in \mathbb{B}$. It follows from (2.7) that $\varphi_j \in H(\overline{\mathbb{B}})$ for each $j = 1, \dots, n$. Consequently, $R^{(k)}\varphi_j$ is in $H(\overline{\mathbb{B}})$ and remains bounded on $\overline{\mathbb{B}}$ for any $k \in \mathbb{N}$. Thus,

$$\begin{aligned} M_\varphi^{(1)} &:= \sup_{z \in \mathbb{B}_n} |R\varphi(z)| < \infty, \\ M_\varphi^{(2)} &:= \sup_{z \in \mathbb{B}_n} |R^{(2)}\varphi(z)| < \infty. \end{aligned} \quad (2.8)$$

Let $\lambda \in (0, 1)$ be such that $|R\varphi(z)| \leq 1$ and $|R^{(2)}\varphi(z)| \leq 1$ for $|\varphi(z)| \leq \lambda$. There exists $D_0 > 0$ such that

$$1 \leq D_0 I_\omega^1(\lambda), \quad 1 \leq D_0 I_\omega^2(\lambda). \quad (2.9)$$

Then, there exists $D_1 > 0$ such that

$$\begin{aligned} &\sup_{|\varphi(z)| \leq \lambda} \omega(\varphi(z)) |R\varphi(z)| (1 + I_\omega^1(\varphi(z))) \\ &\leq D_1 \sup_{|\varphi(z)| \leq \lambda} \omega(\varphi(z)) |R\varphi(z)| I_\omega^1(\varphi(z)), \\ &\sup_{|\varphi(z)| \leq \lambda} \omega(\varphi(z)) |R^{(2)}\varphi(z)| (1 + I_\omega^2(\varphi(z))) \\ &\leq D_1 \sup_{|\varphi(z)| \leq \lambda} \omega(\varphi(z)) |R^{(2)}\varphi(z)| I_\omega^2(\varphi(z)). \end{aligned} \quad (2.10)$$

Let $D = \max\{D_0 + 1, D_1\}$. For every $f \in \mathcal{Z}_\omega$, by (2.1)–(2.4), (2.10), and a standard calculation, we have

$$\begin{aligned} &\omega(z) |R^{(2)}(f \circ \varphi)(z)| \\ &\leq \omega(z) [|R^{(2)}f(\varphi(z))\varphi(z)| \\ &\quad + 2|Rf(\varphi(z))R\varphi(z)| + |f(\varphi(z))R^{(2)}\varphi(z)|] \\ &= \frac{\omega(z)}{\omega(\varphi(z))} \omega(\varphi(z)) [|R^{(2)}f(\varphi(z))\varphi(z)| \\ &\quad + 2|Rf(\varphi(z))R\varphi(z)| + |f(\varphi(z))R^{(2)}\varphi(z)|] \\ &\leq \frac{\omega(z)}{\omega(\varphi(z))} \left[1 \right. \\ &\quad + C\omega(\varphi(z)) (2|R\varphi(z)|(1 + I_\omega^1(\varphi(z))) \\ &\quad \left. + |R^{(2)}\varphi(z)|(1 + I_\omega^2(\varphi(z)))) \right] \|f\|_{\mathcal{Z}_\omega} \end{aligned} \quad (2.11)$$

$$\begin{aligned} &\leq \frac{\omega(z)}{\omega(\varphi(z))} \left[1 \right. \\ &\quad \left. + CD \sup_{|\varphi(z)| \geq \lambda} \omega(\varphi(z)) [2M_\varphi^{(1)} + M_\varphi^{(2)}] \right] \|f\|_{\mathcal{Z}_\omega} \\ &= \frac{\omega(z)}{\omega(\varphi(z))} \left[1 + CDR_\omega [2M_\varphi^{(1)} + M_\varphi^{(2)}] \right] \|f\|_{\mathcal{Z}_\omega} \end{aligned}$$

for every $z \in \mathbb{B}$.

(i) We begin by considering the case when $a = 0$. In this situation, we have $|\varphi(z)| \leq |z|$ for all $z \in \mathbb{B}$. Denote

$$B_\delta := \{z \in \mathbb{B} : |\varphi(z)| \leq \delta\}.$$

Since μ is decreasing on $[\delta, 1)$ we have

$$\begin{aligned} \frac{\omega(z)}{\omega(\varphi(z))} &\leq \frac{M_\omega}{m_{\omega,\delta}}, \quad z \in B_\delta; \\ \frac{\omega(z)}{\omega(\varphi(z))} &< 1, \quad z \in \mathbb{B} \setminus B_\delta. \end{aligned}$$

Therefore, it follows from (2.11) that

$$\begin{aligned} &\sup_{z \in \mathbb{B}_n} \omega(z) |R^{(2)}(f \circ \varphi)(z)| \\ &\leq \sup_{z \in B_\delta} \omega(z) |R^{(2)}(f \circ \varphi)(z)| \\ &\quad + \sup_{z \in \mathbb{B}_n \setminus B_\delta} \omega(z) |R^{(2)}(f \circ \varphi)(z)| \\ &\leq \left(\frac{M_\omega}{m_{\omega,\delta}} + 1 \right) \\ &\quad \times \left(1 + CDR_\omega [2M_\varphi^{(1)} + M_\varphi^{(2)}] \right) \|f\|_{\mathcal{Z}_\omega} < \infty. \end{aligned} \quad (2.12)$$

Hence, $f \circ \varphi \in \mathcal{Z}_\omega$.

(ii) Now, we consider the case $a \neq 0$. Take a $\gamma \in \text{Aut}(\mathbb{B})$ such that $\gamma(0) = a$. Then $\eta := \varphi \circ \gamma \in \text{Aut}(\mathbb{B})$ and $\eta(0) = 0$. By (i), $g := f \circ \eta \in \mathcal{Z}_\omega$. Note that $\gamma^{-1} \in \text{Aut}(\mathbb{B})$, as the above, we have $R^{(k)}\gamma^{-1}$ is bounded in $\overline{\mathbb{B}}$ for any positive integer k . Then, since $f \circ \varphi = g \circ \gamma^{-1}$, as the estimate (2.12) we have

$$\begin{aligned} &\sup_{z \in \mathbb{B}_n} \omega(z) |R^{(2)}(f \circ \varphi)(z)| \\ &= \sup_{z \in \mathbb{B}_n} \omega(z) |R^{(2)}(g \circ \gamma^{-1})(z)| \\ &\leq \left(\frac{M_\omega}{m_{\omega,\delta}} + 1 \right) \\ &\quad \times \left(1 + CDR_\omega [2M_{\gamma^{-1}}^{(1)} + M_{\gamma^{-1}}^{(2)}] \right) \|g\|_{\mathcal{Z}_\omega} < \infty. \end{aligned}$$

Consequently, $f \circ \varphi \in \mathcal{Z}_\omega$. $\square$

Remark 2.3. The condition $I_\omega^2(1) < \infty$ cannot be omitted. Indeed, consider the weight function $\omega(t) = (1-t)^2$ for $t \in [0, 1)$ which satisfies $I_\omega^2(1) = \infty$. It is easy to see that the function $f(z) = \ln(1-z)$, which belongs to $\mathcal{Z}_\omega$, does not admit a continuous extension to $\overline{\mathbb{D}}$. This means that the condition (i) is not true for $\mathcal{Z}_\omega$.

3. A RELATION BETWEEN WEIGHTED COMPOSITION OPERATORS $\mathcal{B}_\omega \rightarrow \mathcal{Z}_\omega$ AND $\mathcal{Z}_\omega \rightarrow \mathcal{Z}_\omega$

In order to conclude the paper we establishes the relation between the boundedness, compactness of weighted composition operators from $\mathcal{B}_\omega$ into $\mathcal{Z}_\omega$ and from $\mathcal{Z}_\omega$ into itself.

Before stating the theorem first let us note that for each $j = 1, \dots, n$ the function id_j given by $id_j(z) := z_j$ belongs to $\mathcal{Z}_\omega$. Then, in the case $\psi \in H^\infty(\mathbb{B})$ with $\|\psi\|_\infty \leq 1$ and $W_{\psi,\varphi} : \mathcal{Z}_\omega \rightarrow \mathcal{Z}_\omega$ is compact, $W_{\psi,\varphi}(id_j) = \psi \cdot \varphi_j$ hence, $\theta_j := \psi \cdot \varphi_j \in \mathcal{Z}_\omega$, $j = 1, \dots, n$. For each $m \geq 1$, put

$$\theta^m = (\theta_1^m, \dots, \theta_n^m) := \prod_{k=0}^{m-1} (\psi \circ \varphi^k) \cdot \varphi^m,$$

where $\varphi^0 = id$, and $\varphi^k := \underbrace{\varphi \circ \dots \circ \varphi}_{k \text{ times}}$ for $k \geq 1$.

Theorem 2.3(i) allows us to assume that θ^m are continuous on $\overline{\mathbb{B}}$ for every $m \geq 0$.

Theorem 3.1. Let $\psi \in H^\infty(\mathbb{B})$, $\varphi = (\varphi_1, \dots, \varphi_n) \in S(\mathbb{B})$ and ν, ω be normal weights on $\mathbb{B}$ and $I_\omega^1(1) < \infty$ Then the following are equivalent:

- (1) $W_{\psi,\varphi} : \mathcal{B}_\nu \rightarrow \mathcal{Z}_\omega$ is compact;
- (2) $W_{\psi,\varphi} : \mathcal{B}_\nu \rightarrow \mathcal{Z}_\omega$ is bounded;
- (3) $W_{\psi,\varphi} : \mathcal{Z}_\omega \rightarrow \mathcal{Z}_\omega$ is compact;
- (4) $\psi, \psi \cdot \varphi_j \in \mathcal{Z}_\omega$ for every $j = 1, \dots, n$ and $\|\psi\|_\infty < 1$.

To establish the theorem, we first require several lemmas.

Lemma 3.2. Assume that $\varphi(0) = 0$ and $W_{\psi,\varphi} : \mathcal{Z}_\omega \rightarrow \mathcal{Z}_\omega$ is compact. Then $\|\theta^m\|_\infty \rightarrow 0$.

Proof. Without loss of generality we may assume that $\|\psi\|_\infty \leq 1$. We have two cases to consider:

(i) When $|\psi(0)| = 1$, it follows from Theorem 2.3(i) and the maximum modulus principle that ψ must be identically equal to 1. Then $W_{\psi,\varphi} = C_\varphi$, the composition operator on $\mathcal{Z}_\omega$, and hence, the lemma follows from Lemma 2.2 of paper.¹⁸

(ii) Now we assume that $|\psi(0)| < 1$.

We will prove that $W_{\psi,\varphi}$ has spectral radius $\varrho(W_{\psi,\varphi}) < 1$.

Let $\lambda \neq 0$ be a spectral point of $W_{\psi,\varphi}$. Since $W_{\psi,\varphi}$ is compact, λ must to be an eigenvalue. Let $f \in \mathcal{Z}_\omega$ be an eigenfunction of $W_{\psi,\varphi}$ corresponding to the eigenvalue λ . Thus $W_{\psi,\varphi}(f) = \lambda f$ and there is a point $a \in \mathbb{B}$ for which $f(a) \neq 0$. Denote $\mathbb{B}^a := \{z \in \mathbb{B} : |z| < \frac{1+|a|}{2}\}$. Note that, $|\varphi(z)| < |z|$ for every $z \in \mathbb{B}$, since otherwise, the composition operator C_φ would be an isomorphism. Consequently, by $\|\psi\|_\infty \leq 1$, $(\psi \cdot C_\varphi)(B_{\mathcal{Z}_\omega})$ is not relatively compact subset of the unit ball $B_{\mathcal{Z}_\omega}$ of $\mathcal{Z}_\omega$. This means $\psi \cdot C_\varphi$ is not a compact operator. This contradicts the compactness of $W_{\psi,\varphi}$. By the Schwarz Lemma, $\varphi(\mathbb{B}^a)$ is relatively compact in $\mathbb{B}^a$. Applying the Schwarz Lemma again to the appropriately normalized restriction of φ on $\varphi(\mathbb{B}^a)$, and continuing this argument, it follows that $\varphi^m(a) \rightarrow 0$ as $m \rightarrow \infty$.

Now, since $\lim_{m \rightarrow \infty} |\psi(\varphi^{m-1}(a))| = |\psi(0)| \neq 1$, by using the fact that, if $0 < a_m < 1$ and $\{a_m\}_{m \geq 1}$ does not converge to 1 then $\prod_{m=1}^\infty a_m = 0$, we obtain

$$\begin{aligned} \lambda^m f(a) &= [W_{\psi,\varphi}]^m(f)(a) \\ &= \left(\prod_{k=0}^{m-1} \psi(\varphi^k(a)) \right) \cdot f(\varphi^m(a)) \rightarrow 0 \cdot f(0) \end{aligned}$$

as $m \rightarrow \infty$. Because $f(a) \neq 0$ it therefore must has $|\lambda| < 1$. The compactness of $W_{\psi,\varphi}$ ensures that its spectrum consists of 0 along with at most countably many eigenvalues accumulating only at 0. Thus, the spectral radius equals the largest eigenvalue in modulus, which, as shown above, is strictly less than 1. It then follows from the spectral radius formula that

$$\lim_{m \rightarrow \infty} \|[W_{\psi,\varphi}]^m\|^{1/m} = \varrho(W_{\psi,\varphi}) < 1,$$

so in particular, $\lim_{m \rightarrow \infty} \|[W_{\psi,\varphi}]^m\| = 0$. Note that $\theta_j^m = [W_{\psi,\varphi}]^m(id_j) \in \mathcal{Z}_\omega$, $j = 1, \dots, n$. Then,

$$\begin{aligned} \|\theta_j^m\|_{\mathcal{Z}_\omega} &= \|[W_{\psi,\varphi}]^m(id_j)\|_{\mathcal{Z}_\omega} \\ &\leq \|[W_{\psi,\varphi}]^m\| \|id_j\|_{\mathcal{Z}_\omega} \rightarrow 0 \quad \text{as } m \rightarrow \infty. \end{aligned}$$

On the other hand, Theorem 2.3(i & iii) shows that $\mathcal{Z}_\omega$ has a topology stronger than that of the sup norm. Hence, $\|\theta^m\|_\infty$ tends to zero as $m \rightarrow \infty$. The lemma is thus proved. $\square$

Lemma 3.3. Suppose $\psi \in H(\mathbb{B})$, $\varphi \in S(\mathbb{B})$, and μ, ν are normal weights on $\mathbb{B}$. Let $X = \mathcal{B}_\nu$ or $\mathcal{Z}_\nu$. Then $W_{\psi,\varphi} : X \rightarrow \mathcal{Z}_\mu$ is compact if and only if, whenever a bounded sequence f_m in X converges to zero uniformly on compact subsets of $\mathbb{B}$, it follows that $\|W_{\psi,\varphi}(f_m)\|_{\mathcal{Z}_\mu} \rightarrow 0$ as $m \rightarrow \infty$.

The lemma for the case $X = \mathcal{B}_\nu$ has been proven in paper.²¹ For the case $X = \mathcal{Z}_\nu$, it is similar to that of $X = \mathcal{B}_\nu$ and will therefore be omitted.

Lemma 3.4. Let $\psi \in H(\mathbb{B})$, $\varphi = (\varphi_1, \dots, \varphi_n) \in S(\mathbb{B})$ and μ, ν be normal weights on $\mathbb{B}$. Assume that $W_{\psi,\varphi} : \mathcal{Z}_\nu \rightarrow \mathcal{Z}_\mu$ is bounded. Then

$$\begin{aligned} \sup_{z \in \mathbb{B}_n} \mu(z) |A_{\psi,\varphi}(z)| &< \infty, \\ \sup_{z \in \mathbb{B}_n} \mu(z) |B_{\psi,\varphi}(z)| &< \infty, \end{aligned} \quad (3.1)$$

where

$$\begin{aligned} A_{\psi,\varphi}(z) &:= 2R\psi(z)R\varphi(z) + \psi(z)R^{(2)}\varphi(z), \\ B_{\psi,\varphi}(z) &:= \psi(z)((R\varphi_1(z))^2, \dots, (R\varphi_n(z))^2). \end{aligned}$$

Proof. First, choosing $f_0(z) = 1 \in \mathcal{Z}_\nu$, the boundedness of $W_{\psi, \varphi}$ implies that $\psi \in \mathcal{Z}_\mu$.

At the same time, for each $j \in \{1, \dots, n\}$, by considering $f_j(z) = z_j$ and $g_j(z) = z_j^2$ for every $z = (z_1, \dots, z_n) \in \mathbb{B}$ we can check that $\psi \cdot \varphi_j, \psi \cdot \varphi_j^2 \in \mathcal{Z}_\mu$.

Then, since

$$\begin{aligned} R^{(2)}[\psi(z)\varphi_j(z)] &= R^{(2)}\psi(z)\varphi_j(z) + 2R\psi(z)R\varphi_j(z) \\ &\quad + \psi(z)R^{(2)}\varphi_j(z) \\ &= R^{(2)}\psi(z)\varphi_j(z) + A_{\psi, \varphi_j}(z), \\ R^{(2)}[\psi(z)\varphi_j^2(z)] &= \varphi_j(z)(R^{(2)}\psi(z)\varphi_j(z) + 4R\psi(z)R\varphi_j(z) \\ &\quad + 2\psi(z)R^{(2)}\varphi_j(z)) + 2\psi(z)(R\varphi_j(z))^2 \\ &= \varphi_j(z)[2R^{(2)}[\psi(z)\varphi_j(z)] - R^{(2)}\psi(z)] \\ &\quad + 2B_{\psi, \varphi_j}(z) \end{aligned} \quad (3.2)$$

for every $z \in \mathbb{B}$ and every $j = 1, \dots, n$ we have

$$\begin{aligned} \sup_{z \in \mathbb{B}_n} \mu(z)|A_{\psi, \varphi_j}(z)| &\leq \|\psi \cdot \varphi_j\|_{\mathcal{Z}_\mu} + \|\psi\|_{\mathcal{Z}_\mu} < \infty, \\ \sup_{z \in \mathbb{B}_n} \mu(z)|B_{\psi, \varphi_j}(z)| &\leq \|\psi \cdot \varphi_j^2\|_{\mathcal{Z}_\mu} + 2\|\psi \cdot \varphi_j\|_{\mathcal{Z}_\mu} + \|\psi\|_{\mathcal{Z}_\mu} < \infty \end{aligned}$$

for every $j = 1, \dots, n$. Consequently, (3.1) is proved. $\square$

Proof of Theorem 3.1. Theorem is trivial if $\|\psi\|_\infty = 0$. Without loss of generality we may assume that $0 < \|\psi\|_\infty \leq 1$, since for otherwise we can consider the $\|\psi\|_\infty^{-1}\psi$ instead of ψ .

(1) $\Rightarrow$ (2): It is obvious.

(2) $\Rightarrow$ (3): Assume that $\{f_m\}_{m \geq 1}$ is a bounded sequence in $\mathcal{Z}_\omega$ which converges to zero uniformly on compact subsets of $\mathbb{B}$. Then, by the Weierstrass theorem, both $\{Rf_m\}_{m \geq 1}$ and $\{R^{(2)}f_m\}_{m \geq 1}$ converge uniformly to zero on compact subsets of $\mathbb{B}$ as well. We will prove that $\|f_m\|_{\mathcal{B}_\omega}$ converges to zero. Given any $\varepsilon > 0$, since $\omega(t) \rightarrow 0$

as $t \rightarrow 1$, we can choose $\varrho \in (\delta, 1)$ so that $\omega(|z|) < \varepsilon$ whenever $\varrho < |z| < 1$. In addition, there exists an integer N such that for all $m \geq N$, we have $|f_m(0)| < \varepsilon$, $|Rf_m(z)| < \varepsilon$, and $|R^{(2)}f_m(z)| < \varepsilon$ for all $|z| \leq \varrho$. Therefore, by (2.3),

$$\begin{aligned} \|f_m\|_{\mathcal{B}_\omega} &\leq |f_m(0)| + \sup_{z \in \mathbb{B}_n} \omega(z)|Rf_m(z)| \\ &\leq \varepsilon + \sup_{|z| \leq \varrho} \omega(z)|Rf_m(z)| \\ &\quad + \sup_{\varrho < |z| < 1} \omega(z)|Rf_m(z)| \\ &\leq \varepsilon + \varepsilon M_\omega + \sup_{\varrho < |z| < 1} \omega(z) \left| Rf_m\left(\frac{z}{2|z|}\right) \right. \\ &\quad \left. + \int_{1/(2|z|)}^1 R^{(2)}f_m(tz) \frac{dt}{t} \right| \\ &\leq \varepsilon + \varepsilon M_\omega + \varepsilon \sup_{m \geq 1} \sup_{|w|=1/2} |Rf_m(w)| \\ &\quad + 2\varepsilon \int_{1/(2|z|)}^{\varrho/|z|} |R^{(2)}f_m(tz)| |z| dt \\ &\quad + 2 \sup_{\varrho < |z| < 1} \omega(z) \int_{\varrho/|z|}^1 |R^{(2)}f_m(tz)| |z| dt \\ &\leq \varepsilon + \varepsilon M_\nu + \varepsilon \sup_{m \geq 1} \sup_{|w|=1/2} |Rf_m(w)| \\ &\quad + 2\varepsilon \|f_m\|_{\mathcal{Z}_\omega} \int_{1/2}^{\varrho} \frac{dt}{\omega(t)} \\ &\quad + 2\|f_m\|_{\mathcal{Z}_\omega} \sup_{\varrho < |z| < 1} \omega(z) \int_{\delta}^{|z|} \frac{dt}{\omega(t)} \\ &\leq \varepsilon + \varepsilon M_\omega + \varepsilon \sup_{m \geq 1} \sup_{|w|=1/2} |Rf_m(w)| \\ &\quad + 2\varepsilon \|f_m\|_{\mathcal{Z}_\omega} \int_{1/2}^{\varrho} \frac{dt}{\omega(t)} + 2\varepsilon \|f_m\|_{\mathcal{Z}_\omega} \\ &\leq \varepsilon K \quad \text{for all } m \geq N. \end{aligned}$$

Then, the boundedness of $W_{\psi, \varphi}$ implies that $\|W_{\psi, \varphi}(f_m)\|_{\mathcal{Z}_\omega} \lesssim \|f_m\|_{\mathcal{B}_\omega} \rightarrow 0$ as $m \rightarrow \infty$. Therefore, $W_{\psi, \varphi}$ is compact by Lemma 3.3.

(3) $\Rightarrow$ (4): Without loss of generality, we may assume that $\|\psi\|_\infty < 1$.

(i) We first consider the case where $\varphi(0) = 0$. Suppose, for the sake of contradiction, that $\|\varphi\|_\infty = 1$. Then there is a rotation $\zeta, z \mapsto e^{i\alpha}z$, such that $\tilde{\varphi} := \zeta \circ \varphi$ has a fixed point $z_0 \in \mathbb{B}_n$. We may choose ζ such that

$\psi(z_0) \neq 0$. Put

$$\tilde{\psi} := \frac{\psi}{\psi(z_0)}.$$

Then, for every $m \geq 1$ we obtain that

$$(\tilde{\theta})^m := \left(\prod_{k=0}^{m-1} \tilde{\psi} \circ (\tilde{\varphi})^k \right) (\tilde{\varphi})^m$$

has a fixed point z_0 , hence, $\|(\tilde{\theta})^m\|_\infty \geq 1$. It follows from Lemma 3.2 that the operator $W_{\tilde{\psi}, \tilde{\varphi}}$, and hence, $W_{\psi, \varphi}$ is cannot be compact.

Note that $W_{\psi, \tilde{\varphi}} = W_{\psi, \varphi} \circ C_\zeta$ where the composition operator C_ζ is an isomorphism of $\mathcal{Z}_\omega$. This implies that $W_{\psi, \varphi}$ is not compact. This contradicts the hypothesis.

(ii) We now consider the case $\varphi(0) = a \neq 0$. Let γ be the conformal automorphism of $\mathbb{B}_n$ taking a to 0, and set $\eta = \gamma \circ \varphi$. It follows from Theorem 2.3(iv & iii) that C_γ is a bounded operator on $\mathcal{Z}_\omega$, hence, $W_{\psi, \eta}$ is compact on $\mathcal{Z}_\omega$ because $W_{\psi, \eta} = W_{\psi, \varphi} \circ C_\gamma$. Finally, it follows from the case (i) that $\|\eta\|_\infty < 1$, and hence, $\|\varphi\|_\infty < 1$.

(4) $\Rightarrow$ (1): Suppose $\{f_m\}_{m \geq 1}$ is a bounded sequence in $\mathcal{B}_\nu$ converging uniformly to zero on compact subsets of $\mathbb{B}$. Using the Cauchy integral formula again, we see that

$$\begin{aligned} \sup_{|\varphi_k(z)| \leq \lambda} |R_{\varphi(z)} f_m(\varphi(z))| &\rightarrow 0, \\ \sup_{|\varphi_k(z)| \leq \lambda} |\nabla_{\varphi(z)}^{(2)} f_m(\varphi(z))| &\rightarrow 0 \quad \text{as } m \rightarrow \infty. \end{aligned}$$

with $\lambda = \|\varphi\|_\infty < 1$, Then, by $\psi \in \mathcal{Z}_\omega$, (3.1) and a standard calculation, we have

$$\begin{aligned} &\|W_{\psi, \varphi}(f_m)\|_{\mathcal{Z}_\omega} \\ &\leq |f_m(0)| + \omega(z) |R^{(2)}[\psi(z)] f_m(\varphi(z))| \\ &\quad + \omega(z) |A_{\psi, \varphi}(z) |R f_m(\varphi(z))| \\ &\quad + \omega(z) |B_{\psi, \varphi}(z) |R^{(2)} f_m(\varphi(z))| \\ &\leq |f_m(0)| + \|\psi\|_{\mathcal{Z}_\omega} \sup_{|\varphi(z)| \leq \|\varphi\|_\infty} |f_m(\varphi(z))| \\ &\quad + \sup_{z \in \mathbb{B}_n} \omega(z) |A_{\psi, \varphi}(z)| \sup_{|\varphi(z)| \leq \|\varphi\|_\infty} |R f_m(\varphi(z))| \\ &\quad + \sup_{z \in \mathbb{B}_n} \omega(z) |B_{\psi, \varphi}(z)| \sup_{|\varphi(z)| \leq \|\varphi\|_\infty} |R^{(2)} f_m(\varphi(z))| \\ &\rightarrow 0 \end{aligned}$$

as $m \rightarrow \infty$. By Lemma 3.3, $W_{\psi, \varphi}$ is compact. $\square$

Acknowledgments

This research is conducted within the framework of science and technology projects at institutional level of Quy Nhon University under the project code T2025.881.01.

REFERENCES

1. S. Stević. On α -Bloch spaces with Hadamard gaps, *Abstract and Applied Analysis*, **2007**, 2007, 39176.
2. C. C. Cowen, B. D. Maccluer. *Composition operators on spaces of analytic functions*, CRC Press, New York, 1995.
3. E. Abbasi, H. Vaezi. Generalized weighted composition operators from the Bloch-type spaces to the weighted Zygmund spaces, *Filomat*, **2019**, 33(3), 981-992.
4. E. Abbasi, X. Zhu. Product-type operators from the Bloch space into Zygmund-type spaces, *Bulletin of the Iranian Mathematical Society*, **2022**, 48, 385-400.
5. B. Choe, H. Koo, W. Smith. Composition operators on small spaces, *Integral Equations Operator Theory*, **2006**, 56, 357-380.
6. Q. Hu, S. Ye. Weighted composition operators on the Zygmund spaces, *Abstract and Applied Analysis*, **2012**, 2012, 462482.
7. H. Li, X. Fu. A new characterization of generalized weighted composition operators from the Bloch space into the Zygmund space, *Journal of Function Spaces and Applications*, **2013**, 2013, 925901.
8. S. Li, S. Stević. Generalized composition operators on Zygmund spaces and Bloch type spaces, *Journal of Mathematical Analysis and Applications*, **2008**, 338, 1282-1295.
9. X. Zhu, N. Hu. Weighted composition operators from Besov Zygmundtype spaces

- into Zygmund-type spaces, *Journal of Function Spaces*, **2020**, 2020, 2384971.
10. J. Du, S. Li, Y. Zhang. Essential norm of generalized composition operators on Zygmund type spaces and Bloch type spaces, *Annales Polonici Mathematici*, **2017**, 119, 107-119.
 11. J. Du, S. Li, Y. Zhang. Essential norm of weighted composition operators on Zygmund-type spaces with normal weight, *Mathematical Inequalities & Applications*, **2018**, 21, 701-714.
 12. Q. Hu, S. Li, Y. Zhang. Essential norm of weighted composition operators from analytic Besov spaces into Zygmund type spaces, *Journal of Contemporary Mathematical Analysis*, **2019**, 54 (3), 129-142.
 13. Y. Liu, Y. Yu. Products of composition, multiplication and radial derivative operators from logarithmic Bloch spaces to weighted-type spaces on the unit ball, *Journal of Mathematical Analysis and Applications*, **2015**, 423, 76-93.
 14. Y. Liu, J. Zhou. On an operator $M_u R$ from mixed norm spaces to Zygmundtype spaces on the unit ball, *Complex Analysis and Operator Theory*, **2013**, 7, 593-606.
 15. S. Li, X. Zhang. Composition operators on the normal weight Zygmund spaces in high dimensions, *Journal of Mathematical Analysis and Applications*, **2020**, 487, 124000.
 16. J. Zhou, Y. Liu. Products of radial derivative and multiplication operator between mixed norm spaces and Zygmund-type spaces on the unit ball, *Mathematical Inequalities & Applications*, **2014**, 17, 349-366.
 17. X. Zhang, S. Xu. Weighted differentiation composition operators between normal weight Zygmund spaces and Bloch spaces in the unit ball of $\mathbb{C}^n$ for $n > 1$, *Complex Analysis and Operator Theory*, **2019**, 13, 859-878.
 18. J. Shapiro. Compact composition operators on spaces of boundary-regular holomorphic functions, *Proceedings of the American Mathematical Society*, **1987**, 100 (1), 121-133.
 19. X. Tang. Extended Cesàro operators between Bloch-type spaces in the unit ball of $\mathbb{C}^n$, *Journal of Mathematical Analysis and Applications*, **2007**, 326, 1199-1211.
 20. K. Zhu. Spaces of holomorphic functions in the unit Ball. *Graduate Texts in Mathematics*, Springer-Verlag, New York, 2005.
 21. L. V. Lam, T. T. Quang. On the boundedness and compactness of extended Cesàro composition operators between weighted Bloch-type spaces, *Hacettepe Journal of Mathematics and Statistics*, **2024**, 53 (4), 897-914.



© 2025 by the authors. This Open Access Article is licensed under the Creative Commons Attribution-NonCommercial 4.0 International (CC BY-NC 4.0) license (<https://creativecommons.org/licenses/by-nc/4.0/>).

Nghiên cứu tối ưu quá trình gia tốc động cơ không đồng bộ ba pha

Đỗ Văn Cần^{1,*}, Thân Văn Thông¹, Hoàng Văn Hiếu²

¹Khoa Kỹ thuật và Công nghệ, Trường Đại học Quy Nhơn, Việt Nam

²Học viên cao học, Khoa Kỹ thuật và Công nghệ, Trường Đại học Quy Nhơn, Việt Nam

Ngày nhận bài: 01/05/2025; Ngày sửa bài: 05/08/2025;

Ngày nhận đăng: 22/08/2025; Ngày xuất bản: 28/10/2025

TÓM TẮT

Bài báo này trình bày nghiên cứu tăng/giảm tốc (gia tốc) của động cơ không đồng bộ ba pha khi khởi động và dừng nhằm nâng cao chất lượng hiệu quả truyền động cho động cơ. Dựa trên điều khiển tốc độ và mômen, tác giả xây dựng quỹ đạo tăng tốc S bậc cao nhằm khắc phục các hạn chế của gia tốc tuyến tính như hiệu suất thấp, rung lắc, giật và ảnh hưởng đến tuổi thọ hệ thống. Các kết quả mô phỏng cho thấy rằng việc sử dụng quỹ đạo S không chỉ giúp giảm độ giật mà còn hạn chế dao động mômen, từ đó cải thiện rõ rệt độ êm ái và hiệu suất hệ truyền động. Ngoài ra, thuật toán xây dựng quỹ đạo S trong gia tốc đã được hiện thực hóa trên phần mềm và cấu hình trên nền tảng phần cứng FPGA. Kết quả thử nghiệm mô phỏng phần mềm, phần cứng cho thấy tín hiệu điều khiển tốc độ được tạo ra với độ phân giải và thời gian đáp ứng cao, phù hợp để tích hợp vào các hệ thống truyền động của động cơ không đồng bộ.

Từ khóa: Động cơ không đồng bộ, truyền động, quá trình gia tốc, quỹ đạo S, FPGA.

*Tác giả liên hệ chính.

Email: dovancan@qnu.edu.vn

Study on optimal acceleration control of three phase induction motors

Do Van Can^{1,*}, Than Van Thong¹, Hoang Van Hieu²

¹*Faculty of Engineering and Technology, Quy Nhon University, Vietnam*

²*Graduate Student, Faculty of Engineering and Technology, Quy Nhon University, Vietnam*

Received: 01/05/2025; Revised: 05/08/2025;

Accepted: 22/08/2025; Published: 28/10/2025

ABSTRACT

This paper presents a study on the acceleration and deceleration process of a three phase induction motor during start up and stop phases, aiming to enhance the dynamic performance of the drive system. Based on torque and speed control principles, the author proposes the application of a high order S-curve acceleration profile to overcome the limitations of linear acceleration, such as reduced efficiency, mechanical vibrations, jerks, and shortened system lifespan. Simulation results demonstrate that the S-curve profile not only reduces jerk but also minimizes torque oscillations, significantly improving the smoothness and overall performance of the drive system. Furthermore, the S-curve profile generation algorithm has been implemented in software and deployed on an FPGA hardware platform. Both software and hardware simulation results confirm that the generated speed control signals achieve high resolution and fast response, making the solution well suited for integration into induction motor drive systems.

Keywords: *Induction motor, drive system, acceleration process, S-curve profile, FPGA.*

1. INTRODUCTION

The acceleration and deceleration process of a three phase induction motor directly affects system efficiency, equipment durability, and the overall lifespan of the drive system. If the acceleration is too slow, it may overload the electrical system and delay motion execution; on the other hand, overly fast acceleration leads to large mechanical forces, vibrations, and early wear.¹ Particularly in elevator applications, sudden changes in speed can cause discomfort, loss of balance, or even health concerns for elderly passengers and children. Therefore, optimizing the acceleration process not only improves operational efficiency and system reliability but also enhances smoothness

and extends mechanical durability in drive applications.

One of the key factors influencing operational quality is the acceleration profile applied during start and stop phases. Currently, linear acceleration profiles are commonly used in inverter based drives; however, they generate high jerk levels at phase transitions, leading to load oscillations, reduced mechanical component lifespan, and degraded user experience.² In contrast, S-curve acceleration allows for much smoother motion, making it better suited for high precision systems such as CNC machines, elevators, and camera positioning systems.

The application of Field Oriented Control (FOC) aims to achieve rapid system stabilization

**Corresponding author.*

Email: dovancaan@qnu.edu.vn

and minimal tracking error, even under variations in motor parameters, load conditions, and external disturbances.³ However, the use of conventional linear acceleration profiles within FOC often results in high jerk, adversely affecting the mechanical load despite the fact that this method remains widely adopted in practice.

FOC and Direct Torque Control (DTC) are the two most common vector control strategies used in electric motor drive systems. FOC employs linear controllers and pulse-width modulation (PWM) techniques to regulate the fundamental voltage components applied to the stator.⁴ In contrast, DTC is a nonlinear control approach that directly generates voltage vectors without the need for a modulation unit. Studies^{5,6} have presented comparative analyses between these two control strategies. The advantages and limitations of each method are analyzed specifically for two types of electric machines: the induction motor (IM) and the permanent magnet synchronous motor (PMSM). The comparisons are based on several evaluation criteria, including fundamental control characteristics, dynamic performance, parameter sensitivity, and implementation complexity.

In paper,⁷ the author proposed a motor speed control method based on an artificial neural network incorporating an online learning algorithm, aimed at compensating for uncertain parameters and large load variations in the dynamic model of an AC motor. Simulation results conducted in MATLAB demonstrated the high control quality and effectiveness of the proposed method, clearly reflecting its strong adaptability when using an online-trained neural network.

Another study^{8,9} also introduced a solution based on a recurrent fuzzy neural network (RFNN) to address this limitation. Specifically, a traditional PID controller was combined with an RFNN-based supervisory controller to dynamically adjust the system's feedback response.¹⁰ Simulation results showed that, under the same set of parameters, the system

exhibited significant overshoot when using only the PID controller. However, when combined with the RFNN supervisory layer, the overshoot phenomenon was completely eliminated.

In studies,^{11,12} a novel adaptive control scheme based on an improved Feedback Error Learning (FEL) method was proposed to address the Load Frequency Control problem. The FEL strategy incorporates both an intelligent controller and a conventional controller operating in the feedforward and feedback paths, respectively. In this scheme, the conventional feedback controller specifically a PID controller, plays a crucial role in ensuring global asymptotic stability of the system. Simultaneously, an intelligent feedforward controller (INFC) is employed to learn the inverse dynamics of the control system. Once the INFC successfully learns the system inverse, reference signal tracking can be achieved accurately during acceleration.

The problem of acceleration based on S-curve trajectories has been widely investigated, particularly in stepper and servo motors.^{13,14} In such systems, abrupt stopping, sudden starting, or rapid speed changes can cause the rotor to lose synchronization due to sudden changes in input pulses or mismatched pulse frequencies. This may lead to step loss, overshoot, and positional errors during motion. Additionally, strong impacts in the mechanical structure may shorten the system's operational life.

To mitigate these motion errors and improve motor stability, various control strategies have been proposed. Experimental results indicate that gradually reducing the speed of stepper motors significantly improves motion stability. However, if the speed is reduced too much, overall performance is compromised. Therefore, designing motor controllers that incorporate well-defined acceleration and deceleration phases can significantly enhance motion efficiency while ensuring smooth operation of the stepper motor.^{15,16}

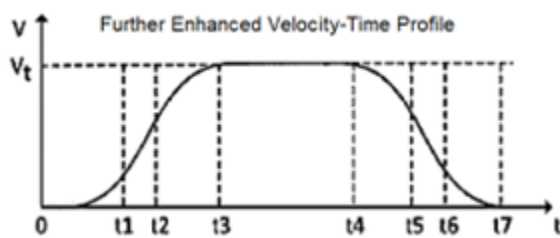


Figure 1. Seven phase S-curve acceleration profile.¹⁷

In addition to servo motors,¹⁸ stepper motors have also been studied for acceleration control in camera motion applications.¹⁹ To ensure accurate camera positioning and maintain visual continuity during motion, smooth speed control of the stepper motor is essential.

In elevator drive systems, induction motors must ensure high stopping accuracy while providing maximum comfort for passengers.²⁰ Moreover, elevators are characterized by frequent start-stop operations, requiring specialized braking systems and reliable speed feedback mechanisms.²¹ The referenced study introduces various types of variable-speed drive systems and how they are applied in electric elevator systems.

Recently, some researchers have proposed jerk-reducing drive strategies for elevator applications.²² However, these studies remain at a fundamental level and have not yet integrated with advanced control strategies for stabilizing both speed and torque.

Thus, previous studies have focused on two main directions: The first direction investigates control methods for induction motors, aiming for speed tracking control that enables the motor speed to quickly respond to the reference speed, without yet addressing the impact of jerk on the load. The second direction explores the acceleration process using S-curve trajectories, with consideration for jerk reduction, but primarily applied to stepper and servo motors in certain applications such as CNC machines, cameras, etc.

While the aforementioned studies focus primarily on achieving fast control response and high tracking accuracy, they often overlook

a critical aspect in practical motor drive applications - the need for a smooth acceleration process during motor start-up, especially for induction motors. Most of these methods aim to minimize steady-state error or adapt to parameter variations through intelligent control strategies such as ANN, RFNN, or FEL-based adaptive schemes. However, they do not adequately address the issue of jerk reduction, which is essential for minimizing mechanical vibrations, reducing wear on mechanical components, and enhancing ride comfort in applications such as elevators or precision motion systems. As a result, despite their effectiveness in dynamic response and robustness, these approaches remain limited in improving the mechanical smoothness and durability of the drive system during the crucial acceleration phase. FOC for controlling speed and torque, combined with high-order S-curve acceleration during startup, helps optimize operational efficiency, reduce power consumption, and extend lifespan and is the chosen solution in this paper.

2. IMPLEMENTATION CONTENT

2.1. Kinematic model of a three phase induction motor

The electromagnetic model of an induction motor is commonly represented in the dq reference frame (rotor flux-oriented), which simplifies torque and speed control when using an inverter. The fundamental equations of this model are as follows:²³

$$\begin{cases} v_d = R_s i_d + \frac{d\psi_d}{dt} - \omega_e \psi_q \\ v_q = R_s i_q + \frac{d\psi_q}{dt} + \omega_e \psi_d \\ \psi_d = L_d i_d + L_m i_{dr} \\ \psi_q = L_q i_q + L_m i_{qr} \end{cases} \quad (1)$$

Where:

i_d, i_q : Stator current in the dq axis (A)

v_d, v_q : Stator voltages in the dq axis (V)

ψ_d, ψ_q : dq axis flux linkages (Wb)

ω_e : Electrical angular speed (rad/s)

R_s : Stator resistance (Ω)

L_d : Inductance (H)

The electromagnetic torque is calculated as follows:

$$M_e = \frac{3}{2} p L_m (i_{qs} i_{dr} - i_{ds} i_{qr}) \quad (2)$$

The inverter is a device that controls the voltage and frequency supplied to an induction motor, enabling precise adjustment of the motor's rotational speed. In motor drive systems, accurate speed control is essential to prevent vibrations during start and stop operations and to maintain stability under varying load conditions. In this study, the authors propose a vector control approach specifically FOC^s combined with the design of an S-curve acceleration profile to enhance motion smoothness and drive performance (Figure 2).

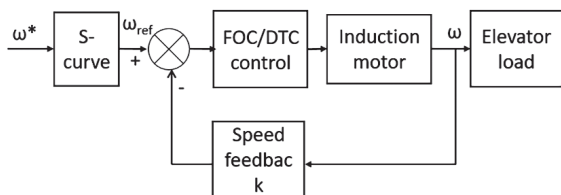


Figure 2. Speed control model with integrated S-curve acceleration profile.

2.2. Developing the acceleration/deceleration algorithm

The jerk equation (3) describes a key factor affecting the acceleration quality of induction motors.

$$j(t) = \frac{da(t)}{dt} = \frac{d^2v(t)}{dt^2} \quad (3)$$

The standard form of this trajectory divides the motion into seven phases, ensuring a continuous change in acceleration (Figure 1): Increasing jerk up to $j_{\max}$; increasing acceleration up to $a_{\max}$; Reducing jerk back to zero $\rightarrow$ maintaining constant velocity; Applying negative

jerk down to $-j_{\max}$; Reducing acceleration back to zero $\rightarrow$ preparing to stop.

An S-curve velocity trajectory is twice differentiable, meaning that both the velocity and acceleration profiles are continuous functions. Let $v(t)$ represent an S-curve velocity trajectory defined over the interval $[0, t]$. Then, for every $t \in [0, t]$, we have:

$$\begin{aligned} v'(t) &= v'(0) + \int_0^t v''(w) dw \\ v(t) &= v(0) + \int_0^t v'(w) dw \end{aligned} \quad (4)$$

The velocity profile $v(t)$ must also satisfy certain boundary conditions. Assuming $v(0) = v_s$ and $v(T) = v_e$ for computational convenience, the profile $v(t)$ can be divided into three segments:

- Acceleration over the time interval $[0, t_a]$, during which the velocity increases from v_s to v_m , where t_a is the acceleration time.
- Deceleration over the time interval $[T - t_d, T]$, during which the velocity decreases from v_m to v_e where t_d is the deceleration time.
- Constant velocity over the time interval $(t_a, T - t_d)$, during which the velocity maintains a fixed value v_m .

Given an initial point and a final point with physical constraints and boundary conditions as described, we set $v''_m = v''_{\max}$ in our proposed S-curve velocity profile to allow acceleration or deceleration to change as rapidly as possible when needed. We then seek an optimal value for v_m under the constraint of total travel distance and subject to the conditions $v_s < v_m \leq v_{\max}$ and $v_e < v_m \leq v_{\max}$, which results in solutions for the acceleration segment, deceleration segment, and constant velocity segment.

The increasing boundary of the S-shaped velocity curve is symmetric about its midpoint $\left(\frac{t_a}{2}, \frac{v_m + v_s}{2}\right)$. Accordingly, the acceleration phase over the time interval $[0, t_a]$ can be subdivided into three subregions corresponding to the previously described stages.

$$\begin{aligned} v(0) &= v_s \\ v(t_a) &= v_m \end{aligned} \quad (5)$$

$$\begin{aligned} v'(0) &= 0 \\ v'(t_a) &= 0 \end{aligned} \quad (6)$$

$$v'(t) = v'_m, t_j \leq t \leq t_a - t_j$$

$$v''(t) = \begin{cases} v''_m, & 0 < t < t_j \\ 0, & t_j \leq t \leq t_a - t_j \\ -v''_m, & t_a - t_j < t < t_a \end{cases} \quad (7)$$

Where v_m , v'_m và v''_m are constants satisfying the conditions: $v_s < v_m < v_{\max}$, $v'_m \leq v'_{\max}$ and $v''_m = v''_{\max}$.

Assuming an initial value v_m is given, along with the corresponding solutions for the acceleration (rising) and deceleration (falling) segments, there are two possible cases concerning the displacement constraint:

Case 1:

$$\int_0^{t_a} v(t)dt + \int_{T-t_d}^T v(t)dt \leq l \quad (8)$$

We have:

$$v_m(T - t_a - t_d) = l - \frac{1}{2}[(v_m + v_s)t_a + (v_m + v_e)t_d] \quad (9)$$

$$T = t_a + t_d + \frac{1}{v_m} \left\{ l - \frac{1}{2}[(v_m + v_s)t_a + (v_m + v_e)t_d] \right\} \quad (10)$$

Case 2:

$$\int_0^{t_a} v(t)dt + \int_{T-t_d}^T v(t)dt > l \quad (11)$$

We must reduce the value of v_m and recalculate the acceleration and deceleration segments to ensure that the displacement constraint is satisfied. The S-curve velocity profile can be solved and optimized using the following binary search method. Given the maximum velocity $v_{\max}$, maximum acceleration $a_{\max}$, and maximum jerk $j_{\max}$, with boundary conditions $v(0) = v_s$, $v(T) = v_e$, t_j , t_a , t_j and t_d the profile components become functions of v_m , and are expressed as follows:

$$t_j = \begin{cases} \sqrt{\frac{v_m - v_s}{v''_{\max}}}, & \sqrt{v''_{\max}(v_m - v_s)} \leq v'_{\max} \\ \frac{v'_{\max}}{v''_{\max}}, & \sqrt{v''_{\max}(v_m - v_s)} > v'_{\max} \end{cases} \quad (12)$$

$$t_a = \begin{cases} 2t_j, & \sqrt{v''_{\max}(v_m - v_s)} \leq v'_{\max} \\ \frac{v_m - v_s}{v'_{\max}} + t_j, & \sqrt{v''_{\max}(v_m - v_s)} > v'_{\max} \end{cases} \quad (13)$$

$$t_{-j} = \begin{cases} \sqrt{\frac{v_m - v_e}{v''_{\max}}}, & \sqrt{v''_{\max}(v_m - v_e)} \leq v'_{\max} \\ \frac{v'_{\max}}{v''_{\max}}, & \sqrt{v''_{\max}(v_m - v_e)} > v'_{\max} \end{cases} \quad (14)$$

$$t_d = \begin{cases} 2t_{-j}, & \sqrt{v''_{\max}(v_m - v_e)} \leq v'_{\max} \\ \frac{v_m - v_e}{v'_{\max}} + t_{-j}, & \sqrt{v''_{\max}(v_m - v_e)} > v'_{\max} \end{cases} \quad (15)$$

The algorithm is constructed as follows:²⁴

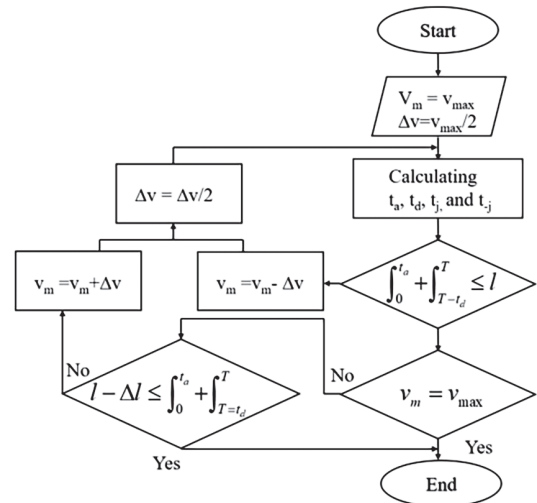


Figure 3. Flowchart of the S-curve trajectory calculation algorithm.

Equations (5) to (7) describe the S-curve velocity trajectory during the acceleration phase using third-order polynomials, ensuring continuity of velocity, acceleration, and jerk. The trajectory is divided into three sub-phases: increasing jerk, maintaining constant acceleration, and decreasing jerk. This structure minimizes mechanical shocks and improves motion smoothness.

Equations (8) to (11) address two cases based on displacement constraints. If the peak

velocity v_{mv_mvm} satisfies the requirement, the total displacement is computed across the three motion phases. Otherwise, v_{mv_mvm} is adjusted, and the segments are recalculated to meet the constraint. Equations (12) to (15) define time and distance for each phase based on the maximum allowable values of velocity, acceleration, and jerk.

The algorithm in Figure 3 applies a binary search method to optimize v_{mv_mvm} , ensuring that the total displacement meets the required condition. The output is a set of smooth, continuous trajectory data points suitable for implementation in digital control systems such as FPGA.

2.3. Integration of S-curve profile with speed control

The S-curve profile generates smooth time-varying references for velocity, position, and torque. Control strategies such as FOC or DTC are responsible for accurately executing the required torque or velocity commands on the motor. Once the FOC controller is in place, it is necessary to generate speed or current (torque) reference signals that precisely follow the S-curve acceleration profile to control the motor accordingly.

The velocity profile $\omega_{ref}(t)$ is pre-generated as a time-base function. It is then compared with the actual motor speed $\omega_{act}(t)$, and a speed PI controller is used to regulate the current specifically, the torque producing current component i_q^*

$$\omega_{ref}(t) = a_0 + a_1 t + a_2 t^2 + a_3 t^3 + a_4 t^4 + a_5 t^5 \quad (16)$$

The coefficients a_i are calculated from the boundary conditions.

$$\begin{aligned} \omega(0) &= 0, \quad \omega(T) = \omega_{max}, \\ \omega'(0) &= \omega'(T) = 0, \quad \omega''(0) = \omega''(T) = 0 \\ i_q^*(t) &= PI[\omega_{ref}(t) - \omega_{act}(t)] \end{aligned} \quad (17)$$

The FOC controller is responsible for converting input signals $i_q^*(t)$ into generated torque.

Direct generation of the reference torque $T_{ref}(t)$: The required acceleration is calculated $a(t) = \dot{\omega}_{ref}(t)$ from the S-curve trajectory, and then the necessary torque is determined accordingly.

$$T_{ref}(t) = J.a(t) \quad (18)$$

$$e(t) = \omega_{ref}(t) - \omega_{act}(t) \quad (19)$$

$$T_{PI}(t) = K_p e(t) + K_i \int e(t) dt \quad (20)$$

$$T_{total}(t) = T_{PI}(t) + T_{ref}(t) \quad (21)$$

$$i_q^*(t) = \frac{2}{3} \frac{T_{total}(t)}{p.\Psi_r} \quad (22)$$

Where:

p : Number of pole pairs

Ψ_r : Rotor flux

The actual torque T_{act} (or the torque-producing current component i_q) is continuously compared with the reference torque $T_{ref}(t)$, and i_q is adjusted accordingly to ensure accurate torque tracking. The S-curve trajectory generator provides a time-based reference for either speed or torque. This reference is then used by the motor control system typically based on FOC or DTC to ensure the motor follows the desired S-curve profile with high precision.

3. SIMULATION RESULTS AND DISCUSSION

3.1. Simulation parameters

Consider a specific case in which a three-phase squirrel-cage induction motor is used for elevator applications and driven through an inverter-motor drive system. Suppose we intend to implement the control strategy using a microcontroller integrated with the inverter.

Table 1. Simulation parameter table.

Parameter	Value
Rated voltage	380 V
Rated frequency	50 Hz
Rated power	3.7 kW
Rated current	7.6 A
Number of poles	4 poles
Rated speed	1445 rpm
Total moment of inertia J	0.045 kg·m ²
Stator resistance R ₁	0.80 Ω
Rotor resistance (referred) R ₂ '	0.85 Ω
Stator inductance L ₁	0.0055 H
Rotor inductance L ₂	0.0055 H
Magnetizing inductance L _m	0.140 H

3.2. Linear acceleration

The linear acceleration process is performed using the motor parameters provided above.

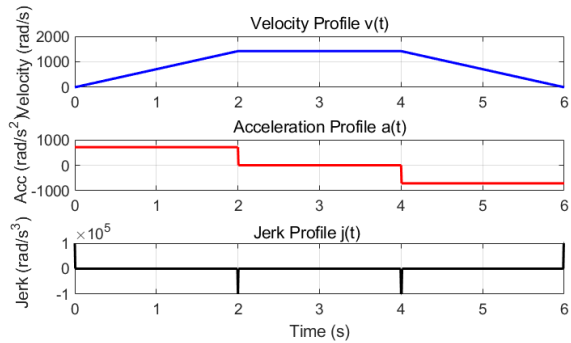


Figure 4. Linear acceleration simulation results.

The simulation results (Figure 4) of the linear acceleration and deceleration process for a 3.7 kW induction motor show that: The motor velocity increases linearly from 0 to its rated speed (1445 rpm) over a 2-second period, remains constant for another 2 seconds, and then decreases linearly back to zero in the final 2 seconds.

During the acceleration phase, the angular acceleration (a) remains constant and positive; it becomes zero during the constant-speed phase, and remains constant and negative during deceleration.

The jerk is approximately zero throughout the uniform acceleration and deceleration phases, except at the phase transitions → specifically at

the end of acceleration (transition to constant speed) and the end of constant speed (transition to deceleration) → where sudden changes in acceleration produce large spikes in jerk.

The most significant jerk spikes are observed at the following transition points:

At time $t = t_{acc} = 2$ seconds: the acceleration phase ends and transitions into the constant-speed phase.

At time $t = t_{acc} + T_{hold} = 4$ seconds: the constant-speed phase ends and transitions into the deceleration phase.

At these two critical points, the acceleration abruptly shifts from a positive value during acceleration to zero at the start of the constant-speed phase, and from zero to a negative value at the start of deceleration. As a result, jerk spikes appear in the plots with extremely high magnitudes, resembling ideal impulse signals with almost instantaneous transitions.

This behavior indicates that acceleration changes instantaneously, with no transitional smoothing phase. As a consequence, both the motor and mechanical load are subjected to significant impulsive forces at the phase transitions.

These high jerk values cause mechanical shock on the motor shaft, drivetrain, and gear mechanisms, increasing the risk of premature failure. The sharp forces also introduce vibrational stresses, which reduce the service life of bearings, gears, and elevator cables. Furthermore, such impulsive dynamics may excite the mechanical system's resonant modes, resulting in unwanted vibrations. From a control perspective, the sudden demand for current and torque adjustment may lead to overshoot, thermal stress in the motor, and even overloading of the inverter system.

3.3. Higher-order acceleration

The velocity trajectory takes the form (16). Boundary conditions: $v(0) = 0$, $v(T) = v_{max}$ (initial, final velocity), $a(0) = 0$, $a(T) = 0$ (initial, final

acceleration 0), $j(0) = 0, j(T) = 0$ (initial, final jerk 0).

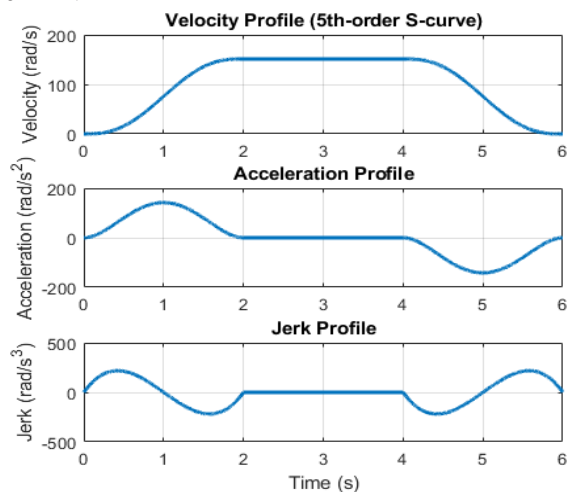


Figure 5. S-curve acceleration for the motor.

The simulation results (Figure 5) of the acceleration constant speed deceleration process of a 3.7 kW motor following a 5th-order S-curve trajectory demonstrate that the velocity increases and decreases smoothly and continuously over time.

In contrast to the constant acceleration observed in linear profiles, the acceleration here progressively increases to a maximum value before smoothly decreasing back to zero.

Jerk does not exhibit sharp spikes as seen in linear acceleration profiles; instead, it varies smoothly with mild oscillations during phase transitions, ensuring higher-order continuity of the trajectory.

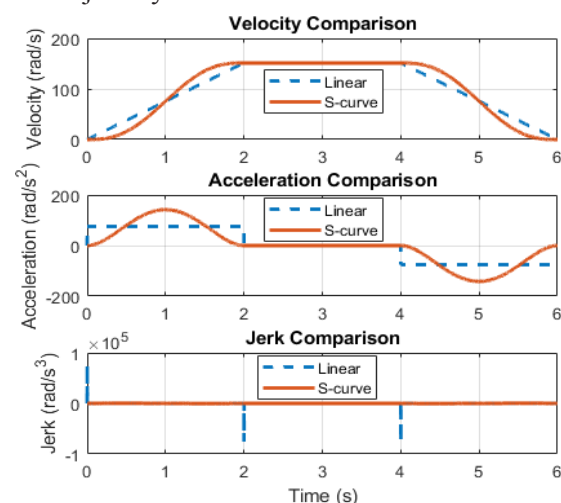


Figure 6. Comparison between linear acceleration and S-Curve acceleration.

The simulation results (Figure 6) of the acceleration, constant speed, and deceleration phases of a 3.7 kW motor using two control strategies linear acceleration and 5th-order S-curve acceleration highlight significant differences in velocity, acceleration, and jerk profiles.

Both methods achieve the target speed (1445 RPM) within the same total duration. However, the velocity, acceleration, and jerk trajectories differ markedly in terms of continuity and smoothness.

Moment 1: End of acceleration phase ($t = t_{acc}$) In the case of linear acceleration/ deceleration, the acceleration drops abruptly from a positive value to zero, resulting in a large jerk spike at the phase transition. This can cause mechanical shocks to the system and sudden inrush currents in the motor. In contrast, the 5th-order S-curve trajectory allows the acceleration to gradually decrease to zero before the end of the acceleration phase. Jerk varies smoothly without large spikes, leading to a gentle transition with no vibration or shock.

Moment 2: End of constant-speed phase, beginning of deceleration ($t = t_{acc} + t_{hold}$) For the linear profile, the acceleration changes abruptly from zero to a negative value, producing another large negative jerk spike. This sudden change may impose unexpected forces on mechanical components, increasing the risk of material fatigue or failure. With the S-curve profile, acceleration decreases smoothly from zero to its negative peak value. Jerk remains continuous, ensuring a soft deceleration phase and improving system durability.

Using linear acceleration, the mechanical system is subjected to abrupt stress at each phase transition due to high jerk, leading to material fatigue and reduced equipment lifespan. In contrast, the 5th-order S-curve ensures continuous changes in acceleration and jerk, distributing forces more evenly throughout the motion. This reduces mechanical shock,

extends the lifespan of the motor, transmission components, and mechanical structure, and significantly enhances system smoothness and operational stability.

3.4. Integrated speed control with S-curve-based acceleration profile

S-curve velocity profile: $\omega(t)$ (rad/s); compute the angular acceleration $a(t)$ and jerk $j(t)$; calculate the reference torque $T(t)$ based on the moment of inertia J :

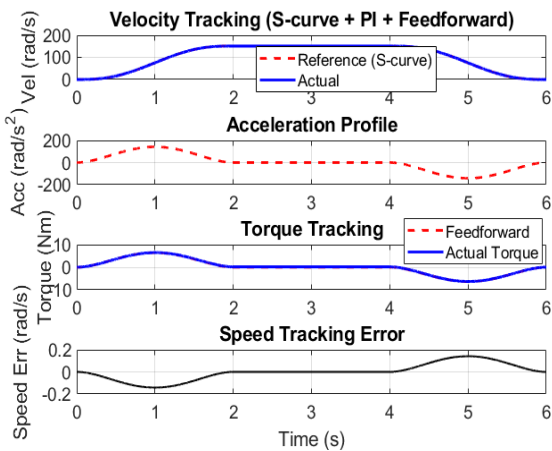


Figure 7. Torque control combined with S-curve acceleration

The simulation results (Figure 7) demonstrate that the combination of S-curve acceleration trajectory planning and torque control using a PI controller with feedforward compensation achieves highly effective speed regulation. The actual motor speed closely follows the reference trajectory over time, with minimal deviation virtually eliminated even at phase transitions between acceleration, constant speed, and deceleration.

The motor torque is adjusted smoothly and continuously thanks to the feedforward torque component, which compensates based on the trajectory acceleration. This allows the motor to respond quickly to required velocity changes without relying entirely on PI feedback. As a result, the system achieves not only high tracking accuracy but also reduces the control burden on the feedback loop, minimizing overshoot and oscillation.

Compared to the model using a standalone PI controller, the addition of feedforward clearly yields smoother, more precise operation and significantly shortens the transient response time. This control strategy is well-suited for high-performance motion systems requiring precision and smoothness, such as premium elevators, industrial robots, or autonomous guided vehicles (AGVs).

3.5. Simulation of FPGA hardware implementation

The `step_out` signal generates pulses with a frequency that varies according to this profile, specifically: the maximum speed is 1445 RPM (approximately 151.3 rad/s), acceleration time is 2 seconds, the peak acceleration is around 200 rad/s², and the peak jerk is approximately 500 rad/s³.

Parameter	value
CLK_PERIOD	20 ns
jerk_val	1000
accel_ticks	5000
start	1 sau 200 ns
sim_time	10 ms

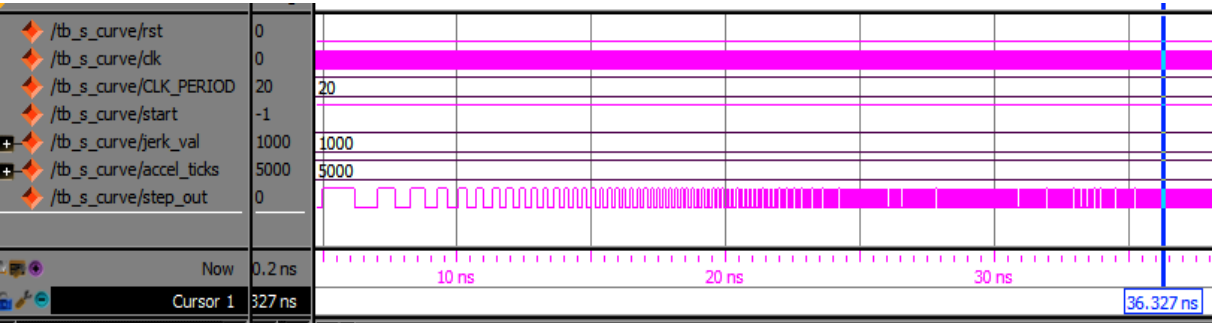


Figure 8. S-curve trajectory acceleration process on FPGA output pin.

The simulation results (Figure 8) show that the `step_out` signal appears with gradually increasing frequency, reflecting a smooth acceleration process characteristic of an S-curve trajectory.

`jerk_val` (unit: Δacc per clock cycle): Controls the rate of acceleration change. A higher value results in faster velocity increase.

`accel_ticks` (number of clock cycles): Defines the total acceleration time. The larger this value, the slower and smoother the acceleration phase becomes.

`step_out`: Output pulse signal with frequency proportional to motor velocity. Observation shows that the pulses become

increasingly dense over time consistent with the expected behavior of S-curve motion.

The simulation confirms that the system operates stably and without glitches, making it suitable for motor control applications requiring high precision and smoothness.

To evaluate the combined effect of S-curve acceleration/deceleration with speed and torque control during the initial 2-second period, the authors performed a scaled simulation in ModelSim using a 2 ms acceleration phase, followed by 2 ms of constant speed, and 2 ms of deceleration. This allows for easier verification and visualization within the simulation environment.

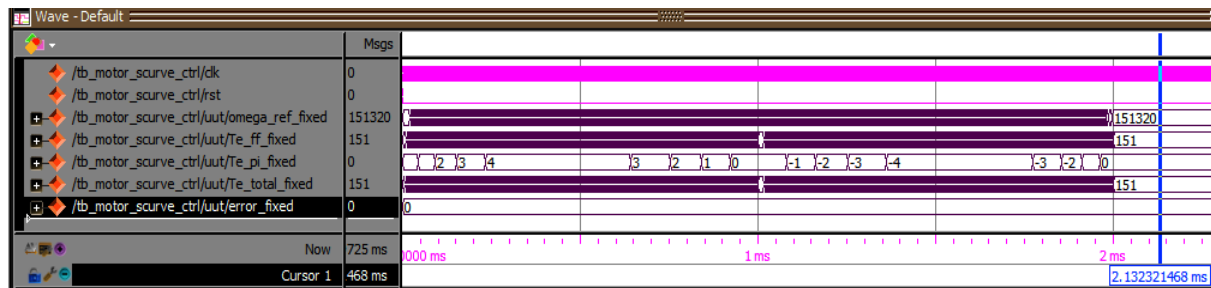


Figure 9. S-curve-based acceleration integrated with speed control.

The simulation results (Figure 9) confirm the effectiveness of integrating a 5th-order S-curve motion profile with a combined PI and feedforward torque control strategy. The system achieves smooth velocity transitions, minimal tracking error, and continuous torque responses without overshoot or delay. The use of fixed-point quantization for velocity, acceleration, and torque signals makes the control scheme well-suited for FPGA implementation, enabling real-time processing and hardware efficiency.

The entire acceleration–deceleration process over a 4-second interval was accurately simulated on ModelSim and verified to be stable, responsive, and glitch-free. This validated model is ready for deployment in high-precision motion control applications, such as elevator drives, industrial automation, and autonomous vehicle motors using FPGA platforms.

3.6. Hardware Implementation on FPGA

The proposed trigonometric S-curve profile was implemented on the DE1-SoC board (FPGA model 5CSEMA5F31) to validate real-time performance. A Verilog-based design generates a one-shot PWM signal, with pulse width modulated via a lookup table (LUT) stored in a .mif file. A time counter compares with the LUT output to produce smooth PWM modulation. The system was tested at the Chip Design Laboratory, Quy Nhon University, with the PWM output available on GPIO_0.

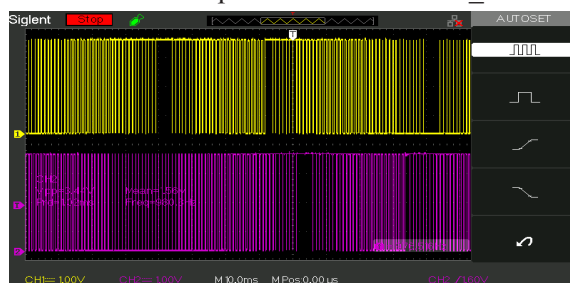


Figure 10. Experimental results obtained on the FPGA in the laboratory.

Compared to paper⁹ where the control system applies linear acceleration and shows significant overshoot, high jerk, and speed ripples during transient states, our method demonstrates superior smoothness and dynamic stability. In¹³ although FOC is implemented, the trajectory generation is limited to basic ramp profiles, lacking jerk control, which causes mechanical stress in high-inertia systems. Meanwhile,²⁵ presents a theoretical multi-segment S-curve trajectory planner but does not apply it to real-time motor control or validate it on embedded platforms.

4. CONCLUSION

This paper has presented an effective approach for implementing smooth and precise motion control by integrating a 5th-order S-curve trajectory with PI-based torque control enhanced by feedforward compensation. Simulation results confirm that the proposed method ensures smooth velocity transitions, continuous acceleration and jerk profiles, and stable torque responses, significantly improving motion quality and system stability.

The control strategy was successfully modeled and verified in ModelSim, where key motion parameters such as speed, acceleration, and torque were quantized using fixed-point representation. This design choice makes the solution highly suitable for real-time implementation on FPGA platforms. The system demonstrated glitch-free behavior, consistent pulse generation (`step_out`), and accurate speed tracking within a compact simulation window, confirming the feasibility of hardware deployment.

The main contributions of this work are:

Development of a 5th-order S-curve motion generator suitable for digital hardware implementation;

Integration of PI and feedforward speed control for improved tracking performance and response smoothness;

Verification of real-time behavior using a hardware-oriented simulation on ModelSim for optimized FPGA resource utilization.

Overall, the proposed model provides a practical and scalable foundation for implementing smooth acceleration and torque control in embedded motion systems using FPGA technology

REFERENCES

1. A. A. Menaem, S. Beryozkina, M. Safaraliev. Intelligent current-speed soft-starting controller for an induction motor drive system, *Journal of Energy Systems*, **2024**, 8(4), 221-236.
2. H. Chen, C. Bi. Optimal starting frequency of three-phase induction motor, *IET Electric Power Applications*, **2022**, 16(3), 362-369.
3. L. T. T. Phuong, L. T. T. Huyen, P. T. H. Anh. Application of rotor-flux-oriented vector control for asynchronous (induction) motors, *Journal of Science & Technology of Technical Universities*, **2017**, 172(1), 115-119.
4. D. V. Can, T. V. Thong. Proposed FPGA-based active disturbance rejection controller (ADRC) for Servo drive systems, *Quy Nhon University Journal of Science*, **2018**, 12(3), 91-102.
5. M. A. W. Begh, H. G. Herzog. Comparison of field oriented control and direct torque control, *Robotics and Control Systems*, **2018**, 1-16.
6. W. J. Xu. Permanent magnet synchronous motor with linear quadratic speed controller, *Energy Procedia*, **2012**, 14(6), 364-369.
7. K. Sabahi, M. Teshnehlab, M. A. Shoorhedeli. Recurrent fuzzy neural network by using feedback error learning approaches for LFC in interconnected power system, *Energy Conversion and Management*, **2009**, 50(4), 938-946.
8. A. A. Ali, F. B. Salem, J. A. K. Mohammed. Design of an electric elevator drive with high riding quality under jerk control, *Engineering*,

- Technology & Applied Science Research*, **2024**, 14(5), 16438-16443.
9. D. V. Can. Current controller design based on FPGA, *Quy Nhon University Journal of Science*, **2020**, 15(1), 71-78.
 10. D. H. D. Khoa, S. H. Thanh, N. C. Ngon. Intelligent control of induction motor using recurrent fuzzy neural networks, *TNU Journal of Science and Technology*, **2022**, 227(08), 46-55.
 11. F. Allam, Z. Nossair, H. Gomma, I. Ibrahim M. Abdelsalam. Evaluation of using a recurrent neural network (RNN) and a fuzzy logic controller (FLC) in closed loop system to regulate blood glucose for type-1 diabetic patients, *International Journal of Intelligent Systems and Applications*, **2012**, 10, 58-71.
 12. D. V. Can, N. V. Linh, T. V. Thong. Research on the design of control system for servo drive systems, *Quy Nhon University Journal of Science*, **2016**, 2(X), 23-30.
 13. B. Kumar, P. Wang, A. S. Hossain. Design of motion profile of stepper motor for elevator using arduino uno, *North American Academic Research Journal*, **2020**, 3(04), 683-695.
 14. A. Drumea, C. I. Marghescu, P. Pantazică, G. Jitianu, A. Vlad. *S-curve motion control implementation using 32-bit microcontroller*, Proceedings of 2022 International Conference on Hydraulics and Pneumatics - HERVEX, Băile Govora, Romania, 2022.
 15. Z. Zhang, Y. Yu. S-type speed control curve based on the number of pulses, *Journal of Physics: Conference Series*, **2022**, 2196(1), 1-10.
 16. V. Kombarov, V. Sorokin, O. Fojtů, Y. Aksonov, Y. Kryzhyvets. S-curve algorithm of acceleration/deceleration with smoothly-limited jerk in high-speed equipment control tasks, *MM Science Journal*, **2019**, 2019(04), 3264-3270.
 17. C. Li, F. Chen. Application of S-curve acceleration and deceleration in winding of bond-wire, *International Journal of Trend in Research and Development*, **2021**, 8(4), 158-161.
 18. A. Z. Mansoor, M. R. Khalil, O. A. Jasim. Position control of dc servo motors using soft-core processor on FPGA to move robot arm, *Journal of Theoretical and Applied Information Technology*, **2011**, 32, 99-106.
 19. X. Wei. Acceleration and deceleration control design of step motor based on TMS320F240, *Procedia Engineering*, **2011**, 15, 501-504.
 20. V. I. Vlachou, D. E. Efstathiou, T. S. Karakatsanis. Design, analysis and application of control techniques for driving a permanent magnet synchronous motor in an elevator system, *Machines*, **2024**, 12(560), 1-33.
 21. R. Akçelik, M. Besley. *Acceleration and deceleration models*, Conference of Australian Institutes of Transport Research (CAITR 2001), Australia, 2002.
 22. M. Osama, O. A. Azim. *Implementation and performance analysis of an elevator electric motor drive system*, 12th International Middle East Power System Conference, Egypt, 2008.
 23. D. V. Can, T. X. Khoa. *Research design position controller based on FPGA*, The 7th International Conference and Exhibition on Control and Automation (VCCA-2024), Hai Phong, Vietnam, 2024.
 24. X. Li. Optimizing S-curve velocity for motion control, *Journal of Applied Mathematics*, **2010**, 1-12.
 25. K. D. Nguyen, T. C. Nguyen, I. M. Chen. On algorithms for planning S-curve motion profiles, *International Journal of Advanced Robotic Systems*, **2008**, 5(1), 99-106.



© 2025 by the authors. This Open Access Article is licensed under the Creative Commons Attribution-NonCommercial 4.0 International (CC BY-NC 4.0) license (<https://creativecommons.org/licenses/by-nc/4.0/>).

Hàm năng lượng tự do của mô hình Ising với từ trường mạnh

Nguyễn Tông Xuân*

Khoa Toán và Thống kê, Trường Đại học Quy Nhơn, Việt Nam

Ngày nhận bài: 08/06/2025; Ngày sửa bài: 19/08/2025;

Ngày nhận đăng: 22/08/2025; Ngày xuất bản: 28/10/2025

TÓM TẮT

Trong bài báo này, chúng tôi trình bày kết quả về miền giải tích của hàm năng lượng tự do cho mô hình Ising, trong đó mỗi spin chỉ tương tác với các spin lân cận của nó. Nghiên cứu của chúng tôi dựa trên khai triển cụm, một công cụ mạnh mẽ trong vật lý thống kê, kết hợp những hiểu biết mới từ Fernandez và Procacci về tiêu chí hội tụ của nó.

Từ khóa: Mô hình Ising, khai triển cụm, hàm năng lượng tự do.

*Tác giả liên hệ chính.

Email: nguyentongxuan@qnu.edu.vn

The free energy function of Ising model in a strong magnetic field

Nguyen Tong Xuan*

Department of Mathematics and Statistics, Quy Nhon University, Vietnam

Received: 08/06/2025; Revised: 19/08/2025;

Accepted: 22/08/2025; Revised: 28/10/2025

ABSTRACT

In this paper, we study the analytic domain of free energy function for Ising model, specifically for cases where only neighboring spins interact. Our study comes from using cluster expansion, a powerful tool in statistical physics, incorporating new insights from Fernandez and Procacci on its convergence criteria.

Keywords: *Ising model, cluster expansion, free energy function.*

1. INTRODUCTION

The Ising model is a mathematical representation of ferromagnetism in statistical mechanics. It consists of discrete variables that represent the magnetic dipole moments of atomic “spins”, which can take on one of two states: +1 or -1. These spins are arranged in a graph, typically a crystal lattice, where the local structure repeats periodically in all directions, allowing each spin to interact with its neighbors. One area of interest for mathematicians and physicists is the analytical validity of the free energy function in the Ising model. These issues can be referenced in Friedli and Velenik’s book.¹

The free energy function in mathematics and physics is defined as the logarithm of the partition function. Analyzing the properties of the free energy function is crucial for understanding phase transitions, particularly in the Ising model. This approach is also applicable to more complex mathematical models in statistical mechanics, such as the Potts model, Blume-Capel model, and Curie-Weiss model. To investigate the analytical properties of the free energy function, we typically calculate its analytical domain.

One of the tools used to study the analytical properties of the free energy function is the cluster expansion technique. The cluster expansion represents a power series with respect to auxiliary parameters, often referred to as fugacity. The first application of the cluster expansion was to examine the pressure function at equilibrium as a power series based on the density function derived from the empirical descriptions of gases and liquids. In the theoretical framework of fluids, these parameters correspond to pressures determined respectively from the ideal gas law or the pressure of certain suitable reference components. This problem is discussed in various references.^{1,2}

Cluster expansion has extensive applications in various fields, including probability theory and improving the bounds on colored graphs. These applications hinge on representing the partition function specific to each problem, along with the associated reference parameters for their characteristics. Further insights into the applications of cluster expansion can be found in references.¹⁻⁴

A common question that arises in these contexts is when the free energy function behaves as an analytic function in relation to the identified parameters.

*Corresponding author.

Email: nguyentongxuan@qnu.edu.vn

One method to improve the analytic range of the free energy function in the Ising model involves in refining the convergence criteria of the cluster expansion.

Study on the convergence domain of cluster expansion began in the late sixties,^{5,6} but it did not receive significant attention until several decades later. The convergence of cluster expansion has been examined using various methods, including Kirkwood-Salzburg equation,⁵ tree-graph boundaries,⁷ induction methods,⁸ and partition schemes⁹ (for a comparison of these methods, see reference¹⁰). Among these approaches, the partition scheme has yielded the most promising results and has been the focus of several applications and improvements in the paper.⁹ This work has been expanded upon in various studies, summarized in reference.² Nevertheless, the advancements made by Fernandez-Procacci regarding the convergence criteria remain the most promising for practical applications.

This paper aims to explore the expansion of the analytical domain of the free energy function for the Ising model, building on the advancements made by Fernandez-Procacci in the convergence domain of the cluster expansion. Specifically, we will provide the analytic domain when the external field is not zero (in reference,¹ this model is referred to as the Ising model with a strong field).

2. ISING MODEL IN A STRONG FIELD AND MAIN RESULTS

To start this section, let us revisit the definition of distances,

$$d(i, j) := \|i - j\|_\infty = \max_{1 \leq k \leq d} |i_k - j_k| \quad (1)$$

for $i, j \in \mathbb{Z}^d$, and

$$d(S, S') := \inf\{d(\kappa, \ell) : \kappa \in S, \ell \in S'\}$$

for $S, S' \subset \mathbb{Z}^d$.

We consider the set $\Omega = \{-1, 1\}^{\mathbb{Z}^d}$. Configurations denote by $\sigma = (\sigma_x)_{x \in \mathbb{Z}^d}$. Let us consider a finite set $\Lambda \subset \mathbb{Z}^d$, configurations $\sigma_\Lambda \in \{-1, 1\}^\Lambda :=$

Ω_Λ and Hamiltonians with free boundary condition

$$H_{\Lambda; \beta, h}^\varnothing(\sigma_\Lambda) := -\beta \sum_{\{i, j\} \in \Lambda} f(i, j) \sigma_i \sigma_j - h \sum_{i \in \Lambda} \sigma_i, \quad (2)$$

where $\beta \in \mathbb{R}_{\geq 0}$ is the inverse temperature, $h \in \mathbb{R}$ is the external field, and the interaction $f(\cdot, \cdot)$ is defined as

$$f(i, j) = \begin{cases} 1 & \text{if } \|i - j\|_\infty = 1 \\ 0 & \text{otherwise} \end{cases}. \quad (3)$$

Consider a configurations $\sigma_\Lambda \omega_{\Lambda^c} \in \Omega$ and Hamiltonians

$$H_{\Lambda; \beta, h}^\omega(\sigma_\Lambda \omega_{\Lambda^c}) := H_{\Lambda; \beta, h}^\varnothing(\sigma_\Lambda) - \beta \sum_{\substack{i \in \Lambda \\ j \in \Lambda^c}} f(i, j) \sigma_i \omega_j, \quad (4)$$

where the interaction $f(\cdot, \cdot)$ is defined in (3), a configuration $\sigma_\Lambda \omega_{\Lambda^c} \in \Omega$ includes two parts $\sigma_\Lambda \in \{-1, 1\}^\Lambda := \Omega_\Lambda$, and $\omega_{\Lambda^c} \in \{-1, 1\}^{\Lambda^c}$ which is usually called a boundary of the system, or configurations are frozen outside of the finite set Λ , and the term

$$\beta \sum_{\substack{i \in \Lambda \\ j \in \Lambda^c}} f(i, j) \sigma_i \omega_j,$$

refers to the interaction between the internal and external components of the system.

The partition function with free boundary condition in Λ is

$$Z_\Lambda^\varnothing(\beta, h) := \sum_{\sigma_\Lambda \in \Omega_\Lambda} \exp\left(-H_{\Lambda; \beta, h}^\varnothing(\sigma_\Lambda)\right), \quad (5)$$

the (finite-volume) free energy function (pressure function) with free boundary condition is

$$P_\Lambda^\varnothing(\beta, h) := \frac{1}{|\Lambda|} \log Z_\Lambda^\varnothing(\beta, h). \quad (6)$$

And the partition function with ω -boundary condition in Λ is

$$Z_\Lambda^\omega(\beta, h) := \sum_{\sigma_\Lambda \in \Omega_\Lambda} \exp\left(-H_{\Lambda; \beta, h}^\omega(\sigma_\Lambda \omega_{\Lambda^c})\right), \quad (7)$$

the (finite-volume) free energy function with ω -boundary condition in Λ is

$$P_\Lambda^\omega(\beta, h) := \frac{1}{|\Lambda|} \log Z_\Lambda^\omega(\beta, h). \quad (8)$$

The thermodynamic free energy function $p^\#$ is obtained through the thermodynamic limit

$$p^\#(\beta, h) = \lim_{\Lambda \uparrow \mathbb{Z}^d} P_\Lambda^\#(\beta, h) \quad (9)$$

in Fisher sense, where $\# := \emptyset$ or ω .

We utilize the fact that thermodynamic pressure is independent of boundary conditions (for reference, please take a look at Theorem 3.8¹) and, for the sake of algebraic convenience, we will focus in this section on “plus” boundary conditions: $\omega_i = 1$ for all $i \notin \Lambda$. The interaction between the inside and outside of the system can be described as the following term

$$\beta \sum_{\substack{i \in \Lambda, j \notin \Lambda \\ \|i-j\|_\infty=1}} \sigma_i \omega_j = \beta \sum_{\substack{i \in \Lambda, j \notin \Lambda \\ \|i-j\|_\infty=1}} \sigma_i.$$

Note that from this point forward, we will use the $+$ notation to represent the plus boundary condition for all previously mentioned quantities. This includes the free energy function in a finite domain, partition functions, and the thermodynamic free energy function.

To get the presentation of partition function, we add and subtract 1 to each term in this Hamiltonian and for each $\sigma_\Lambda \in \Omega_\Lambda$, let us introduce the set

$$\Lambda^-(\sigma_\Lambda) = \{i \in \Lambda : \sigma_i = -1\}. \quad (10)$$

We obtain

$$H_{\Lambda; \beta, h}^+(\sigma_\Lambda) = -\beta |\mathcal{E}_\Lambda| - h |\Lambda| + 2\beta |\partial_e \Lambda^-(\sigma_\Lambda)| + 2h |\Lambda^-(\sigma_\Lambda)| \quad (11)$$

where

$$\partial_e \Lambda^-(\sigma_\Lambda) = \left\{ \{i, j\} : i \in \Lambda^-(\sigma_\Lambda), j \notin \Lambda^-(\sigma_\Lambda), \|i-j\|_\infty = 1 \right\}$$

and

$$\mathcal{E}_\Lambda = \{ \{i, j\} \subset \mathbb{Z}^d : \{i, j\} \cap \Lambda \neq \emptyset, \|i-j\|_\infty = 1 \}.$$

Each σ_Λ corresponds one-to-one to a term of deviation from the ground state $\Lambda^-(\sigma_\Lambda)$ (the configuration

with minimal energy), which is the “all +1” configuration. As a consequence, the partition function can be expressed in terms of deviations from the ground state:

$$Z_\Lambda^+(\beta, h) = \exp(\beta |\mathcal{E}_\Lambda| + h |\Lambda|) Z_\Lambda^{\text{LF}}(\beta, h), \quad (12)$$

where the “large field” polymers partition function $Z_\Lambda^{\text{LF}}(\beta, h)$ is given as

$$Z_\Lambda^{\text{LF}}(\beta, h) := \sum_{\Lambda^- \subset \Lambda} \exp(-2\beta |\partial_e \Lambda^-| - 2h |\Lambda^-|).$$

From the definition of the distance in (1), let us declare that two vertices $i, j \in \Lambda^-$ are *connected* if and only if $d(i, j) \leq 1$, and we can decompose Λ^- into maximally connected components (For example, see Figure 1),

$$\Lambda^- = S_1 \cup \dots \cup S_n$$

with $d(S_\ell, S_k) > 1$ for $\ell \neq k$.

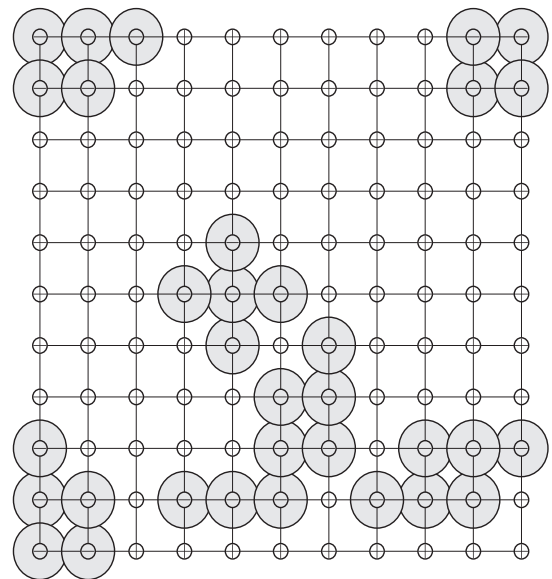


Figure 1. A configuration of the Ising model. Each connected component of the shaded area delimits one of the polymers $S_1, \dots, S_6$.

Before giving an alternative expression of large field polymers partition function, let us introduce the definitions of compatible and incompatible objects as follows:

Definition 2.1. Let us define S, S' to be compatible, and denote $S \sim S'$, if $d(S, S') \geq 2$. Otherwise S , and S' are incompatible and we denote $S \approx S'$.

Denote

$$\zeta(S, S') = \begin{cases} 1 & \text{if } S \sim S' \\ 0 & \text{if } S \approx S' \end{cases}.$$

Since $|\partial_e \Lambda^-| = \sum_{i=1}^n |\partial_e S_i|$ and $|\Lambda^-| = \sum_{i=1}^n |S_i|$, then the expression of large field polymers partition function can be rewritten in the following form

$$Z_{\Lambda}^{\text{LF}}(\beta, h) = 1 + \sum_{n \geq 1} \frac{1}{n!} \sum_{(S_1, \dots, S_n) \in \mathcal{P}_{\Lambda}^n} \prod_{1 \leq i < j \leq n} \zeta(S_i, S_j) \prod_{\ell=1}^n w_{\beta, h}(S_{\ell}), \quad (13)$$

with

$$\mathcal{P}_{\Lambda} = \{S \subset \Lambda : S \text{ is non-empty and connected of } \Lambda\}$$

and

$$w_{\beta, h}(S) = \exp(-2\beta|\partial_e S| - 2h|S|). \quad (14)$$

For $n \in \mathbb{N}$, let $\mathcal{C}[n]$ be the set of all connected graphs on n vertices. For each graph $G = (V(G), E(G))$, let $V(G)$ denote the vertex set of the graph G and $E(G)$ denote the edge set of the graph G .

Theorem 2.1. The pressure with $+$ -boundary condition in Λ can be expressed as the following form

$$P_{\Lambda}^+(\beta, h) = \beta \frac{|\mathcal{E}_{\Lambda}|}{|\Lambda|} + h + \frac{1}{|\Lambda|} \log Z_{\Lambda}^{\text{LF}}(\beta, h), \quad (15)$$

where

$$\begin{aligned} \log Z_{\Lambda}^{\text{LF}}(\beta, h) &= \sum_{n=1}^{\infty} \frac{1}{n!} \sum_{(S_1, \dots, S_n) \in \mathcal{P}_{\Lambda}^n} a_n^T(S_1, \dots, S_n) \prod_{i=1}^n w_{\beta, h}(S_i), \end{aligned} \quad (16)$$

with Ursell function $\omega_n^T(\cdot)$ defined as

$$a_n^T(S_1, \dots, S_n) := \sum_{G \in \mathcal{C}[n]} \prod_{\{i, j\} \in E(G)} [\zeta(S_i, S_j) - 1]. \quad (17)$$

Expression (16) is well-known as cluster expansion.

Proof. Expression (13) serves as the partition function for a gas of polymers, which is comprised of subsets of Λ , as discussed in Fernandez and al. paper.¹⁰ Consequently, we can apply the theory developed for these systems to prove expression (15). For more details, see references.^{10,11} $\square$

The next theorem establishes a sufficient condition for the existence of the pressure function as $\Lambda \rightarrow \mathbb{Z}^d$ in the thermodynamic limit and allows us to verify the analytic domain of the pressure function.

Denote

$$\mathcal{P}_{\mathbb{Z}^d} = \{S \subset \mathbb{Z}^d : S \neq \emptyset \text{ and connected of } \mathbb{Z}^d\},$$

and $\mathcal{P}_{\Lambda} \rightarrow \mathcal{P}_{\mathbb{Z}^d}$ as $\Lambda \rightarrow \mathbb{Z}^d$.

Theorem 2.2. If there exists $a > 0$ such that

$$\begin{aligned} \sum_{k=1}^{\infty} |\mathcal{A}_k| e^{-2d\beta k^{(d-1)/d} V(1)^{1/d} - 2hk + (2d+1)ak} \\ \leq e^a - 1 \end{aligned} \quad (18)$$

with

$$\mathcal{A}_k := \{S \in \mathcal{P}_{\mathbb{Z}^d} : 0 \in S, |S| = k\}, \quad (19)$$

and $V(1)$ denoting the volume of a d -dimensional sphere of radius 1, then the following hold:

(i.) Consider $w_{\beta, h} = \{w_{\beta, h}(S)\}_{S \in \mathcal{P}_{\mathbb{Z}^d}}$. For $S \in \mathcal{P}_{\mathbb{Z}^d}$, the series $|\Gamma|_S(w_{\beta, h})$ defined in (24) is convergent. Furthermore,

$$|\Gamma|_S(w_{\beta, h}) \leq e^{a|S|}.$$

(ii.) The free energy function (15) converges absolutely and uniformly in Λ , and

$$p^+(\beta, h) = \beta d + h + \sum_{X \subset \mathbb{Z}^d : X \ni 0} \frac{1}{|X|} \Psi(X), \quad (20)$$

where, for each $X \subset \mathbb{Z}^d$, $\Psi(\cdot)$ is defined as follow:

$$\begin{aligned} \Psi(X) &= \sum_{n=1}^{\infty} \frac{1}{n!} \sum_{\substack{(S_1, \dots, S_n) \in \mathcal{P}_{\mathbb{Z}^d}^n \\ S_1 \cup \dots \cup S_n = X}} a_n^T(S_1, \dots, S_n) \prod_{i=1}^n w_{\beta, h}(S_i) \end{aligned} \quad (21)$$

Proof. Part (i) of Theorem 2.2 follows from Lemma 4.2, which will be presented in Subsection 4.2 of this paper.

Part (ii) of Theorem 2.2 is derived from the Fernandez-Procacci Theorem.⁹ We will omit the detailed proof and refer readers to^{1,11} for further information. $\square$

The main purpose of this paper is to identify the domain of inverse temperature β and the external magnetic field h for which the pressure function $p(\beta, h)$ is analytic, as stated in the following theorem.

Theorem 2.3. *The pressure function $p(\beta, h)$ is analytic in the domain $\mathcal{D}$ with*

$$\mathcal{D} = \{(\beta, h) \in \mathbb{R} \times \mathbb{R} : \beta \geq 0; 2h \geq \phi_1(\bar{a})\}$$

where ϕ_1 is defined in (47) and $\bar{a}$ in (50).

3. CLUSTER EXPANSION FOR SUBSET GASES

Subset gases are specific types of polymer gases that are frequently utilized in cluster expansion within statistical mechanics. Their definition requires a countable subset, denoted by, $\mathbb{V}$ (e.g. $\mathbb{Z}^d$) which acts as an underlying “space”. Polymers are defined as finite, non-empty subsets of $\mathbb{V}$, represented mathematically as

$$\mathcal{P}_{\mathbb{V}} = \{S \subset \mathbb{V} : 0 < |S| < \infty\}.$$

with compatibility relation, denoted by $S \sim S'$ depending on the models we are working with. For instance, in Section 2, we stated that $S \sim S'$ if and only if $d(S, S') \geq 2$. In the work of Bissacot, Procacci and Fernandez,¹⁰ it is mentioned that $S \sim S'$ if and only if $S \cap S' = \emptyset$. Polymers can now be measured through its cardinality, so it makes sense to talk about large and small polymers. The definition of the gas is completed by a family of activities $\mathbf{z} = \{z_S \in \mathbb{C}\}_{S \in \mathcal{P}_{\mathbb{V}}}$. Let us define the partition function for gas polymers as follow

$$Z(\mathbf{z}) = 1 \quad (22)$$

$$+ \sum_{n \geq 1} \frac{1}{n!} \sum_{(S_1, \dots, S_n) \in \mathcal{P}_{\mathbb{V}}^n} \prod_{1 \leq i < j \leq n} \zeta(S_i, S_j) \prod_{i=1}^n z_{S_i}.$$

Using Mayer’s trick (which can be found in¹¹), we can derive $\log Z(\mathbf{z})$ as the following form

$$\begin{aligned} \log Z(\mathbf{z}) &= \sum_{n \geq 1} \frac{1}{n!} \sum_{(S_1, \dots, S_n) \in \mathcal{P}_{\mathbb{V}}^n} a_n^T(S_1, \dots, S_n) \prod_{i=1}^n z_{S_i}, \end{aligned} \quad (23)$$

where $a_n^T(\cdot)$ is defined in (17).

To study the convergence of cluster expansion, we typically examine it through the convergence conditions of the formal power series in infinite volume as below (see⁹ for a full explanation): For each $S \in \mathcal{P}_{\mathbb{V}}$,

$$\begin{aligned} |\Gamma|_S(\boldsymbol{\rho}) &= 1 \\ &+ \sum_{n=1}^{\infty} \frac{1}{n!} \sum_{(S_1, \dots, S_n) \in \mathcal{P}_{\mathbb{V}}^n} |a_{n+1}^T(S, \dots, S_n)| \prod_{i=1}^n \rho_{S_i} \end{aligned} \quad (24)$$

with $\boldsymbol{\rho} \in [0, \infty)^{\mathcal{P}_{\mathbb{V}}}$.

We will now examine some convergent conditions of cluster expansion as given in equation (24). As a consequence of Theorem 2.3.2,¹¹ the cluster expansion is convergent under Fernandez-Procacci criterion known as the best convergent condition, which is given in the following theorem.

Theorem 3.1 (Fernández-Procacci criterion). *Suppose that for some $\boldsymbol{\xi} \in [0, \infty)^{\mathcal{P}_{\mathbb{V}}}$ there exists $\boldsymbol{\mu} \in [0, \infty)^{\mathcal{P}_{\mathbb{V}}}$ such that*

$$\xi_{S_0} \psi_{S_0}^{\text{FP}}(\boldsymbol{\mu}) \leq \mu_{S_0}, \quad \text{for each } S_0 \in \mathcal{P}_{\mathbb{V}} \quad (25)$$

with

$$\psi_{S_0}^{\text{FP}}(\boldsymbol{\mu}) = 1 + \sum_{n \geq 1} \frac{1}{n!} \sum_{\substack{(S_1, \dots, S_n) \in \mathcal{P}_{\mathbb{V}}^n \\ S_0 \not\sim S_i, S_i \sim S_j, 1 \leq i, j \leq n}} \prod_{i=1}^n \mu_{S_i} \quad (26)$$

Then the series $|\Gamma|_S(\boldsymbol{\xi})$ defined in (24) is convergent. Furthermore, for each $S \in \mathcal{P}_{\mathbb{V}}$,

$$\xi_S |\Gamma|_S(\boldsymbol{\xi}) \leq \mu_S.$$

To apply the Fernández-Procacci criterion, for most of the models, we substitute $\mu_\gamma = \xi_\gamma e^{a|\gamma|}$ to obtain

$$1 + \sum_{n \geq 1} \sum_{\substack{\{S_1, \dots, S_n\} \subset \mathcal{P}_V \\ S_0 \cap S_i \neq \emptyset, S_i \cap S_j = \emptyset, 1 \leq i < j \leq n}} \prod_{i=1}^n \xi_i e^{a|S_i|} \leq e^{a|S_0|}. \quad (27)$$

From the constraint in the sum, $S_0 \cap S_i \neq \emptyset$, $S_i \cap S_j = \emptyset$, $1 \leq i < j \leq n$, this means that each of the polymers $S_1, \dots, S_n$ must intersect different points in S_0 to avoid overlapping. Consequently, we can conclude that: (i) $n \leq |S_0|$, and (ii) there are n different points in S_0 touched by $S_1 \cup \dots \cup S_n$. The selection of these points can be done in $\binom{|S_0|}{n}$ ways. Hence the left-hand side of (27) is less than or equal to

$$1 + \sum_{n=1}^{|S_0|} \binom{|S_0|}{n} \left[\sup_{x \in S_0} \sum_{\substack{S \in \mathcal{P}_V \\ S \ni x}} \xi_S e^{a(S)} \right]^n = \left[1 + \sup_{x \in S_0} \sum_{\substack{S \in \mathcal{P}_V \\ S \ni x}} \xi_S e^{a(S)} \right]^{|S_0|}. \quad (28)$$

This leads us to the following sufficient condition for (27)

$$\sup_{x \in S_0} \sum_{\substack{S \in \mathcal{P}_V \\ S \ni x}} \xi_S e^{a|S|} \leq e^a - 1. \quad (29)$$

This condition, in fact, coincides with the known (but forgotten) Gruber-Kunz condition⁵ except that the later involves a sharp inequality sign. The condition is useful for numerous applications including contour ensembles of low-temperature phases, geometrical objects of high-temperature expansions, random sets of the Fortuin-Kasteleyn representation of the Potts model,...

4. PROOFS

4.1. Alternative Gruber-Kunz condition

In this section, we will utilize the Fernandez-Procacchi criterion given in Section 3 along with a new compatible relation presented in Section 2 to

derive the improvement of Gruber-Kunz condition which is presented in the following proposition.

Proposition 4.1. *If there exists $a > 0$ such that*

$$\sup_{x \in \mathbb{Z}^d} \sum_{\substack{x \in S \\ S \in \mathcal{P}_{\mathbb{Z}^d}}} w_{\beta,h}(S) e^{a|[S]_1|} \leq e^a - 1 \quad (30)$$

with

$$[S]_1 := \{j \in \mathbb{Z}^d : d(j, S) \leq 1\}, \quad (31)$$

then $|\Gamma|_S(w_{\beta,h})$ converges, for $w_{\beta,h} = \{w_{\beta,h}(S)\}_{S \in \mathcal{P}_{\mathbb{Z}^d}}$. Furthermore, for every $S \in \mathcal{P}_{\mathbb{Z}^d}$,

$$|\Gamma|_S(w_{\beta,h}) \leq e^{a|S|}.$$

Proof. Let us start with following conclusion readily from Theorem 3.1 that $|\Gamma|_S(\rho)$ converges if for each $S \in \mathcal{P}$,

$$1 + \sum_{n \geq 1} \sum_{\substack{\{S_1, \dots, S_n\} \in \mathcal{P}_{\mathbb{Z}^d}^n \\ S \sim S_i, S_i \sim S_j, 1 \leq i, j \leq n}} \prod_{i=1}^n w_{\beta,h}(S_i) e^{a(S_i)} \leq e^{a(S)} \quad (32)$$

where we take $\mu_S = w_{\beta,h}(S) e^{a(S)}$. It is easy to see that

$$S \sim S' \iff d(S, S') \leq 1 \iff S \cap [S']_1 = \emptyset$$

with $[S]_1$ defined in (31), and

$$S \sim S' \Rightarrow d(S, S') > 1 \Rightarrow S \cap S' = \emptyset.$$

It implies that the left-hand side of convergent condition (32) can be bounded by

$$1 + \sum_{n \geq 1} \sum_{\substack{\{S_1, \dots, S_n\} \subset \mathcal{P}_{\mathbb{Z}^d} \\ [S]_1 \cap S_i \neq \emptyset, S_i \cap S_j = \emptyset, 1 \leq i, j \leq n}} \prod_{i=1}^n w_{\beta,h}(S_i) e^{a(S_i)}. \quad (33)$$

Then we can replace the convergent condition (32) by

$$1 + \sum_{n \geq 1} \sum_{\substack{\{S_1, \dots, S_n\} \subset \mathcal{P}_{\mathbb{Z}^d} \\ [S]_1 \cap S_i \neq \emptyset, S_i \cap S_j = \emptyset, 1 \leq i, j \leq n}} \prod_{i=1}^n w_{\beta,h}(S_i) e^{a|[S]_1|} \leq e^{a|[S]_1|}. \quad (34)$$

By an argument analogous to that used to derive the Gruber-Kunz condition (29), starting with the constraint in the sum, $[S]_1 \cap S_i \neq \emptyset$, $S_i \cap S_j = \emptyset$, $1 \leq$

$i < j \leq n$, this means that each of the polymers $S_1, \dots, S_n$ must intersect different points in $[S]_1$ to avoid overlapping. Consequently, we can conclude that: (i) $n \leq |[S]_1|$, and (ii) there are n different points in $[S]_1$ touched by $S_1 \cup \dots \cup S_n$. The selection of these points can be done in $\binom{|[S]_1|}{n}$ ways. Hence the left-hand side of (27) is less than or equal to

$$1 + \sup_{x \in \mathbb{Z}^d} \sum_{\substack{x \in S \\ S \in \mathcal{P}_{\mathbb{Z}^d}}} w_{\beta,h}(S) e^{a|[S]_1|} \leq e^a. \quad (35)$$

The proof has been completed. $\square$

4.2. Proof of Theorem 2.3

Before proceeding with further calculations, let us establish a weaker condition for the convergence of the power series $|\Gamma|_S(\cdot)$ in the following lemma. This condition arises from a bound on the weight $w_{\beta,h}(\cdot) e^{a|[S]_1|}$, as outlined in this lemma, along with the Gruber-Kunz condition. This bound is particularly useful for estimating the parameters β and h .

Lemma 4.2. *For each $S \in \mathcal{P}_{\mathbb{Z}^d}$,*

$$w_{\beta,h}(S) \leq e^{-\beta d V_d(1)^{1/d} |[S]|^{(d-1)/d} - h|S|}, \quad (36)$$

with $V(1)$ denoting the volume of a sphere of radius 1, and

$$e^{a|[S]_1|} \leq e^{a(2d+1)|S|}. \quad (37)$$

Furthermore, if there exists $a > 0$ such that

$$\begin{aligned} & \sum_{k=1}^{\infty} |\mathcal{A}_k| e^{-2d\beta k^{(d-1)/d} V(1)^{1/d} - 2hk + (2d+1)ak} \\ & \leq e^a - 1 \end{aligned} \quad (38)$$

with

$$\mathcal{A}_k := \{S \in \mathcal{P}_{\mathbb{Z}^d} : 0 \in S, |S| = k\}, \quad (39)$$

then $|\Gamma|_S(w_{\beta,h})$ converges and

$$|\Gamma|_S(w_{\beta,h}) \leq e^{a|S|},$$

for each $S \in \mathcal{P}_{\mathbb{Z}^d}$.

Proof. We observe that

$$|S| \leq |[S]_1| \leq (2d+1)|S|, \quad (40)$$

then the inequality (37) holds.

To prove the inequality (36), we begin with the fact that the smallest ratio of area to volume is achieved by a d -dimensional sphere. Denoting the volume and surface area of a sphere of radius R , respectively,

$$\begin{aligned} V_d(R) &= V_d(1) R^d \\ S_d(R) &= dV_d(1) R^{d-1} \end{aligned}$$

we obtain

$$\begin{aligned} |\partial_e S| \geq S_d(R) &= dV_d(1)^{1/d} [V_d(R)]^{(d-1)/d} \\ &= dV_d(1)^{1/d} |S|^{(d-1)/d}. \end{aligned} \quad (41)$$

As a consequence of inequalities (36), (37), and the condition (38), we obtain

$$\begin{aligned} & \sup_{x \in \mathbb{Z}^d} \sum_{\substack{x \in S \\ S \in \mathcal{P}_{\mathbb{Z}^d}}} e^{-2\beta|\partial_e S| - 2h|S| + a|[S]_1|} \\ & \leq \sum_{k=1}^{\infty} |\mathcal{A}_k| e^{-2d\beta k^{(d-1)/d} V(1)^{1/d} - 2hk + (2d+1)ak} \\ & \leq e^a - 1 \end{aligned} \quad (42)$$

with $\mathcal{A}_k$ defined in (39). Thus, the alternative Gruber-Kunz condition is satisfied, completing the proof as a consequence of Proposition 4.1. $\square$

The next two lemmas are crucial for determining the number of elements in $\mathcal{A}_k$.

Lemma 4.3 (Lemma 3.60¹). *Let G be connected graph with n edges. Starting from an arbitrary vertex of G , there is a path in G crossing each edge of G exactly twice and ending at the starting vertex.*

Proof. The proof uses an induction argument. For $n = 1$ the result is trivial. Assume that the claim holds for $k = n - 1$ and consider a connected graph G with n -edges. Let i_0 be the starting vertex in G and consider a vertex $j_0 \in V(G)$ such that $\{i_0, j_0\} \in E(G)$. There are two possibilities for $E(G) \setminus \{i_0, j_0\}$:

• It equals $E(G_1)$ where G_1 is a connected graph. In this case, the desired path is obtained by concatenating $\{i_0, j_0\}$ with the path in G_1 starting from j_0 and satisfying the inductive hypothesis, followed by the final step $\{i_0, j_0\}$.

• It equals $E(G_1) \cup E(G_2)$ where G_1 and G_2 are each connected but they are mutually disconnected. The initial site i_0 is a vertex of G_1 and j_0 a vertex of G_2 . In this case the desired path is obtained by the following concatenation: First $\{i_0, j_0\}$, second the inductive path in G_2 starting and ending at j_0 , third $\{i_0, j_0\}$ again, and fourth the inductive path in G_1 starting and ending at i_0 .

In both cases, we obtain a path that starts and ends at i_0 , with exactly two visits to each edge of G . $\square$

Lemma 4.4. For $k \geq 1$,

$$|\mathcal{A}_k| \leq (2d)^{2k-2}. \quad (43)$$

where $\mathcal{A}_k$ is defined in (39).

Proof. For each set $S \in \mathcal{P}_{\mathbb{Z}^d}$ such that $0 \in S$ and $|S| = k$, there exists a spanning tree that contains $k-1$ edges. Consequently, by applying Lemma (4.3), the number of connected sets with k elements that include 0 is bounded above by the number of paths of length $2k-2$ starting from 0. This quantity is certainly less than $(2d)^{2k-2}$. $\square$

We now start with the proof of our main result in this paper. The infinite volume free energy $p(\beta, h)$ inherits the analyticity of the positive series $|\Gamma|_S(\cdot)$ (for more details, see references^{1,11}). Thus we can confirm that all values of the external field h that meet the condition (38) are sufficient for the analyticity of $p(\beta, h)$.

Proof of Theorem 2.3. We start with the condition (38), and apply Lemma 4.4 to obtain

$$\begin{aligned} & \sum_{k=1}^{\infty} |\mathcal{A}_k| e^{-2d\beta V(1)^{1/d} k^{(d-1)/d} - 2hk + (2d+1)ak} \\ & \leq \sum_{k=1}^{\infty} (2d)^{2k-2} e^{[-2h + (2d+1)a]k} \end{aligned}$$

$$= \frac{e^{(2d+1)a-2h}}{1 - (2d)^2 e^{(2d+1)a-2h}}, \quad (44)$$

for $h > 0$ satisfying

$$4d^2 e^{(2d+1)a-2h} < 1. \quad (45)$$

Inequality (44) implies that (38) is valid for h whenever there exists a constant $a > 0$ such that

$$\frac{e^{(2d+1)a-2h}}{1 - (2d)^2 e^{(2d+1)a-2h}} \leq e^a - 1 \quad (46)$$

It is equivalent to

$$\begin{aligned} 2h & \geq \ln[1 + 4d^2(e^a - 1)] - \ln(e^a - 1) + 2da + a \\ & := \phi_1(a) \end{aligned} \quad (47)$$

and from the condition (45), we have

$$2h > 2\ln(2d) + 2da + a := \phi_2(a).$$

Then, for each $a > 0$,

$$2h \geq \max[\phi_1(a), \phi_2(a)] = \phi_1(a). \quad (48)$$

For $a > 0$, we have

$$\ln[1 + 4d^2(e^a - 1)] - \ln(e^a - 1) \geq \ln(4d^2).$$

Thus, for each $a > 0$, as a consequence of the inequality (48), to optimize the domain for the field h , we can take

$$2h \geq \min_{a>0} \phi_1(a). \quad (49)$$

Elementary analysis shows that $\phi_1(a)$ has a unique minimum at

$$\bar{a} = \ln \left[\frac{(2d+1)(8d^2-1) + 1 + \sqrt{\Delta}}{8d^2(2d+1)} \right], \quad (50)$$

with

$$\sqrt{\Delta} = [(2d+1)(8d^2-1)+1]^2 - 16(2d+1)^2(4d^4-d^2).$$

Hence,

$$2h \geq \phi_1(\bar{a}). \quad (51)$$

The proof of Theorem 2.3 is completed. $\square$

4.3. Comparison with Friedli and Velenik result

In the last part, we compare our estimations with the results provided in Friedli and Velenik's book¹ which is one of the newest result on this topic. To describe the latter, let us denote

$$\eta(h, d) = \sum_{k=1}^{\infty} (2d)^{2k} e^{(2d+1-2h)k}$$

$$H^+ = \{h : \operatorname{Re} h \geq \bar{h}\} \quad (52)$$

with

$$\bar{h} := \inf\{h : \eta(h, d) < 1, h > 0\}.$$

Friedli and Velenik state that if $h \in H^+$, then the cluster expansion in the equation (16) converges absolutely. By performing a simple computation, we can estimate that

$$2\bar{h} = \ln(8d^2) + 2d + 1. \quad (53)$$

As a consequence of inequality (49), plug in $a = \ln 2$ into the function $\phi_1(a)$, we have

$$2\bar{h} = \ln(8d^2) + 2d + 1$$

$$\geq \ln(4d^2 + 1) + d2 \ln 2 + \ln 2 \geq \min_{a>0} \phi_1(a). \quad (54)$$

Inequality (54) shows that our estimate is lower than Friedli and Velenik's bound¹ (for more details, see Figure 2). Let us examine the ratio between our bound and Friedli and Velenik's bound¹, as defined below

$$r(d) := \frac{\phi_1(\bar{a})}{\log(8d^2) + 2d + 1},$$

where ϕ_1 is defined in (47) and $\bar{a}$ in (50). In Figure 3, we observe that the rate R decays exponentially, approaching zero as d tends to infinity.

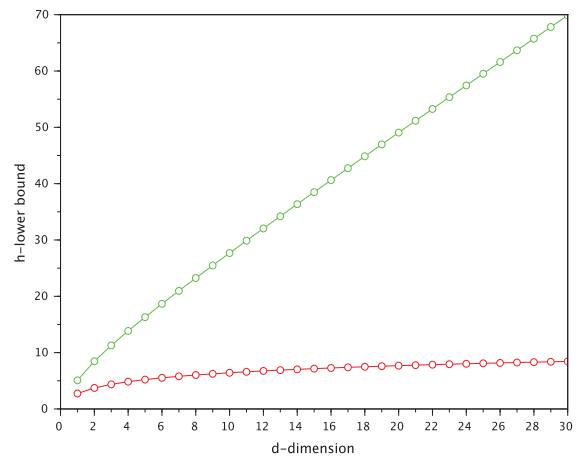


Figure 2. A comparison with Friedli and Velenik result (green line presented for Friedli and Velenik's result and red line presented for our result).

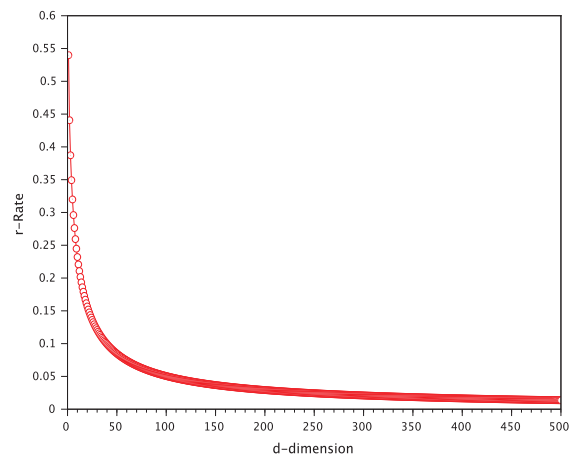


Figure 3. The rate between our bound and Friedli and Velenik bound.

Acknowledgements

The author wishes to express his deepest gratitude to Prof. Roberto Fernández for his helpful suggestions during the preparation of the paper. This research is conducted within framework of science and technology projects at institution level of Quy Nhon University under the project code T2025.882.02.

REFERENCES

1. S. Friedli, Y. Velenik. *Statistical mechanics of lattice systems: a concrete mathematical introduction*, Cambridge University Press, United Kingdom, 2017.
2. N. T. Xuan, R. Fernández. Convergence of cluster and virial expansions for repulsive classical gases, *Journal of Statistical Physics*, **2020**, 179, 448-484.
3. B. Jackson, A. Procacci, A. D. Sokal. Complex zero-free regions at large $|q|$ for multivariate Tutte polynomials (alias Potts-model partition functions) with general complex edge weights, *Journal of Combinatorial Theory Series B*, **2013**, 103, 21-45.
4. S. Ndreca, A. Procacci, B. Scoppola. Improved bounds on coloring of graphs, *European Journal Combinatorics*, **2012**, 33, 592-609.
5. C. Gruber, H. Kunz. General properties of polymer systems, *Communications in Mathematical Physics*, **1971**, 22, 133-161.
6. O. Penrose. Convergence of fugacity expansions for classical systems. *Statistical mechanics: foundations and applications*, W. A. Ben-jamin, New York, 1967.
7. D. C. Brydges. A short cluster in cluster expansions. *Critical Phenomena, Random Systems, Gauge Theories*, Elsevier, North-Holland, 1984.
8. R. L. Dobrushin. Estimates of semiinvariants for the Ising model at low temperatures. *Topics in Statistics and Theoretical Physics*, American Mathematical Society Translations, United States, 1996.
9. R. Fernández, A. Procacci. Cluster expansion for abstract polymer models. New bounds from an old approach, *Communications in Mathematical Physics*, **2007**, 274(1), 123-140.
10. R. Bissacot, R. Fernández, A. Procacci. On the convergence of cluster expansions for polymer gases, *Journal of Statistical Physics*, **2010**, 139, 598-617.
11. N. T. Xuan. *Convergence of cluster and virial expansions. Theory and applications*, Ph.D dissertation, Gran Sasso Science Institute, Italy, 2020.



© 2025 by the authors. This Open Access Article is licensed under the Creative Commons Attribution-NonCommercial 4.0 International (CC BY-NC 4.0) license (<https://creativecommons.org/licenses/by-nc/4.0/>).

Thiết kế anten mảng trạm gốc có độ lợi cao, mức búp sóng bên thấp cho hệ thống thông tin 5G dải tần dưới 6 GHz

Đặng Thị Từ Mỹ, Hồ Văn Phi, Huỳnh Nguyễn Bảo Phương*

Khoa Kỹ thuật và Công nghệ, Trường Đại học Quy Nhơn, Việt Nam

Ngày nhận bài: 05/06/2025; Ngày sửa bài: 12/08/2025;

Ngày nhận đăng: 03/09/2025; Ngày xuất bản: 28/10/2025

TÓM TẮT

Trong bài báo này, một anten mảng có độ lợi cao, mức búp sóng bên thấp được đề xuất cho ứng dụng trạm gốc 5G dưới 6 GHz hoạt động trong băng tần C. Anten này dựa trên thiết kế lưỡng cực điện từ (ME) đã được điều chỉnh, bao gồm hai tấm kim loại hình tam giác, hai tấm kim loại hình chữ nhật, các cột kim loại nối đất, một đầu dò hình L, mặt phẳng đất và đường tiếp điện vi dải. Phần tử anten đơn, với kích thước tổng thể $96\text{ mm} \times 96\text{ mm} \times 2,4\text{ mm}$ (xấp xỉ $1,55\lambda_0 \times 1,55\lambda_0 \times 0,04\lambda_0$ tại 4,9 GHz), đạt băng thông trở kháng rộng từ 4,50 GHz đến 5,06 GHz với hệ số phản xạ ($|S_{11}|$) dưới -10 dB, và độ lợi cực đại 3,1 dBi. Một mảng tuyến tính 1×8 phần tử lưỡng cực ME được phát triển sử dụng kích thích đồng pha và đồng biên. Cấu hình này mang lại độ lợi cực đại cao là 13,8 dBi; tuy nhiên, cũng dẫn đến mức búp sóng bên khá cao là -12,9 dB. Để giải quyết hạn chế này, kỹ thuật giảm biên độ hình thang được áp dụng cho tiếp điện mảng. Các kết quả mô phỏng cho thấy anten mảng cải thiện đáng kể mức búp sóng phụ, đạt dưới -28 dB trong khi vẫn duy trì độ lợi cao 13,6 dBi.

Từ khóa: Độ lợi cao, mức búp sóng bên, anten mảng, thông tin 5G dưới 6 GHz.

*Tác giả liên hệ chính.

Email: huynhnguyenbaophuong@qnu.edu.vn

Design of high gain, low side-lobe level base station array antenna for 5G-sub 6 GHz communication system

Dang Thi Tu My, Ho Van Phi, Huynh Nguyen Bao Phuong*

Faculty of Engineering and Technology, Quy Nhon University, Vietnam

Received: 05/06/2025; Revised: 12/08/2025;

Accepted: 03/09/2025; Published: 28/10/2025

ABSTRACT

In this paper, a high-gain and low side-lobe level array antenna is proposed for 5G sub-6 GHz base station applications operating in the C-band. The antenna is based on a modified magneto-electric (ME) dipole design, comprising two triangular metallic plates, two rectangular metallic plates, grounded vias, an L-shaped probe, a ground plane, and a microstrip feed line. A single antenna element, with overall dimensions of $96 \text{ mm} \times 96 \text{ mm} \times 2.4 \text{ mm}$ (approximately $1.55\lambda_0 \times 1.55\lambda_0 \times 0.04\lambda_0$ at 4.9 GHz), demonstrates a wide impedance bandwidth ranging from 4.50 GHz to 5.06 GHz with a reflection coefficient ($|S_{11}|$) below -10 dB, and achieves a peak gain of 3.1 dBi. A 1×8 linear array of ME dipole elements is developed using uniform in-phase and equal-amplitude excitation. This configuration yields a high peak gain of 13.8 dBi; however, it also results in relatively high side-lobe levels, reaching -12.9 dB. To address this limitation, a trapezoidal amplitude tapering technique is applied to the excitation of the array. The simulated results indicate a significant improvement, achieving a side-lobe level below -28 dB while maintaining a high gain of 13.6 dBi.

Keywords: *High gain, side-lobe level, array antenna, 5G-sub 6 communications.*

1. INTRODUCTION

With the exponential growth of mobile data traffic and the proliferation of smart devices, the evolution of cellular communication technologies toward the fifth generation (5G) has become indispensable. While millimeter-wave (mmWave) technologies have garnered attention due to their large bandwidths, the sub-6 GHz frequency range remains crucial for wide-area coverage, better penetration, and reliable connectivity, especially for 5G macro base station deployments.¹⁻³ Consequently, the design of high-performance antenna arrays suitable for 5G-sub 6 GHz applications has emerged as a vital research focus.

Among various antenna technologies, magneto-electric (ME) dipole antennas have demonstrated significant advantages for base station applications. Formed by combining electric and magnetic dipole components, these antennas offer wide bandwidth, stable gain, and low cross-polarization levels. Their inherently symmetrical structure and differential feeding make them well-suited for dual-polarized arrays, which are essential in modern MIMO systems.⁴⁻⁶ In addition, ME dipoles have also been widely studied for mmWave applications, such as circularly polarized and wideband aperture-coupled arrays at 60 GHz, which further highlight their versatility across different frequency ranges.⁷

*Corresponding author.

Email: huynhnguyenbaophuong@qnu.edu.vn

Recent works have extended ME dipole designs into planar and low-profile configurations by leveraging printed circuit board (PCB) technology to enable mass production and ease of integration. Arrays composed of ME dipoles have demonstrated the ability to maintain stable radiation patterns, support beamforming, and achieve wide-angle scanning-capabilities that are essential for dynamic 5G environments.⁸ However, most studies have predominantly focused on the mmWave bands, whereas optimized designs for the sub-6 GHz spectrum remain relatively underexplored.

In addition to impedance bandwidth and polarization diversity, two essential performance indicators for 5G base station antenna arrays in the sub-6 GHz band are realized gain and side-lobe level (SLL). High gain ensures that the antenna can support long-distance communication and high data throughput, which are crucial for macrocell coverage and enhanced user experiences in dense urban and rural environments. Meanwhile, low SLL is vital to minimize interference, improve beamforming precision, and enhance signal-to-noise ratio in multi-user MIMO systems. Recent investigations on high-gain base station antenna arrays for mmWave cellular communications also demonstrate the importance of carefully optimizing array architectures to achieve both high efficiency and practical deployment feasibility.⁹

Recent studies have demonstrated significant improvements in gain performance of ME dipole arrays designed for sub-6 GHz. For example, the dual-polarized array achieved a peak gain of 9.2 dBi across a wide operating band from 3.3 GHz to 4.2 GHz, while maintaining a compact form factor suitable for 5G base stations.¹⁰ Another study introduced a filtering ME dipole antenna with enhanced radiation performance, demonstrating a peak gain of 10.4 dBi and a front-to-back ratio exceeding 20 dB.¹¹ These characteristics make such designs favorable for sectorized base station deployment.

Furthermore, the control of side-lobe levels has been a focus in optimizing array

radiation patterns. A 4×4 dual-polarized ME dipole array was proposed with a side-lobe suppression of better than -13 dB across the E-plane and H-plane, ensuring minimal leakage of radiated power into undesired directions.¹² The incorporation of parasitic elements and advanced feed networks has also contributed to consistent low SLLs while retaining high efficiency.¹³

Maintaining an optimal balance among gain, SLL, and structural simplicity continues to present a fundamental trade-off. Consequently, ongoing research focuses on techniques such as amplitude tapering, optimization of element spacing, and ground plane shaping to further enhance the radiation characteristics of ME dipole arrays operating in the sub-6 GHz band.^{14,15} These innovations are driven by the need to address practical deployment challenges, including wind resistance, manufacturing cost-efficiency, and seamless integration of the array into compact radome structures. Overall, the continuous evolution in ME dipole array design demonstrates promising potential to deliver the desired electrical and mechanical performance needed for next-generation 5G sub-6 GHz base station antennas.

This study presents a high-gain, high-side-lobe suppression magneto-electric (ME) dipole antenna designed for 5G-sub 6 GHz applications, specifically operating at 4.9 GHz within the C-band spectrum. The antenna is excited using an L-shaped probe, which effectively stimulates the ME dipole mode while preserving a low-profile structure suitable for compact deployments. To evaluate its performance in practical scenarios, a 1 × 8 linear antenna array configuration was implemented and optimized for base station use. Furthermore, by employing trapezoidal excitation techniques, the array achieved a peak gain of 13.6 dBi and exhibited side-lobe levels below -28 dB, indicating excellent radiation pattern control and suitability for high-performance wireless communication systems.

The remainder of the paper is organized as follows. Section 2 provides detailed designs and performance simulations for the single ME dipole antenna. A 1 × 8 elements linear array

design with in phase equal-magnitude and in phase tapering-magnitude feeding are given in Section 3, while some conclusions and future development of are contained in Section 4.

2. SINGLE ANTENNA DESIGN

2.1. Antenna geometry

The antenna's geometry is depicted in Fig. 1. The proposed antenna made up by two dielectric layers which are FR4_epoxy sheet with a relative permittivity of 4.4. The ME dipole is printed on the substrate #1 with a thickness of 1.6 mm and a dimension of $W_{GND} \times W_{GND}$. The antenna consists of four metallic plates, six metallic posts, an L-shaped probe, and a ground plane. Plated through-hole technology was used to create the metallic posts with a diameter of 1.0 mm. A 50 Ohm microstrip line was connected to the vertical portion of the 1.0 mm diameter L-shaped probe. The feeding line was printed on the substrate #2, which has a 0.8 mm thickness.

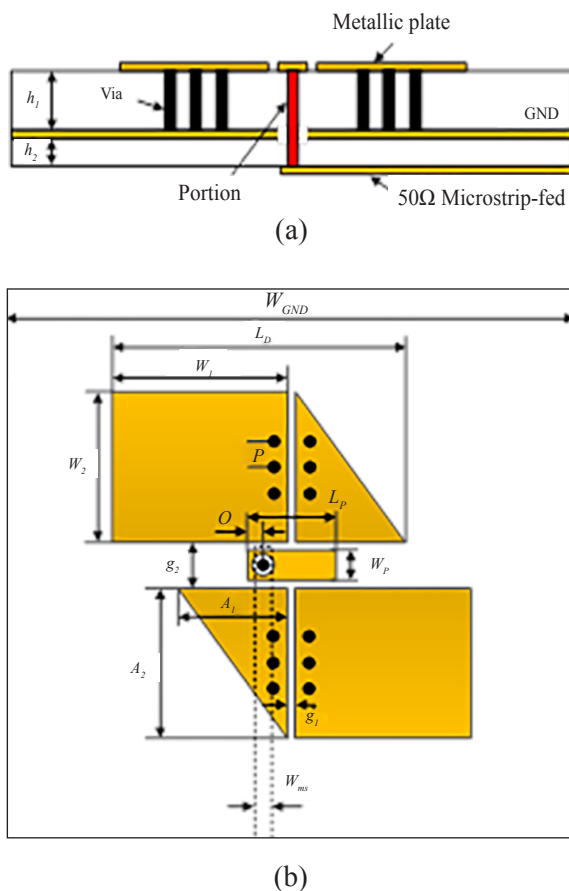


Figure 1. Geometry of the proposed antenna: (a) side view, (b) top view.

2.2. Parameter study

To identify the optimal dimensional parameters of the proposed antenna, it is essential to initially approximate initial values, including the ME dipole length and the width W_{ms} of the 50-ohm feed line. Subsequently, the resonance frequency is examined based on these initial values and then compared to the intended design frequency. The optimization process is then carried out by analyzing the dimensional parameters of the single element antenna. Specifically, each parameter is varied individually to observe its effect on the resonant frequency, while keeping the remaining parameters constant.

Given a specified center resonance frequency, the length L_d of the ME antenna can be estimated. In practice, the ME antenna's length L_d is initially approximated as half of the free-space wavelength at the center design frequency. Hence, the initial estimate of L_d can be readily computed, yielding a value of approximately 30.6 mm (which corresponds to $\sim 0.5\lambda_0$ at 4.9 GHz). The characteristic impedance Z_0 can be calculated with given the dimensions of the microstrip line as paper.¹⁶

$$Z_0 = \begin{cases} \frac{60}{\sqrt{\epsilon_{eff}}} \ln \left[\frac{8h}{w_0} + \frac{w_0}{4h} \right], & \frac{w_0}{h} \leq 1 \\ \frac{120\pi}{\sqrt{\epsilon_{eff}} \left[\frac{w_0}{4h} + 1.393 + 0.667 \ln \left(\frac{w_0}{4h} + 1.444 \right) \right]}, & \frac{w_0}{h} > 1 \end{cases} \quad (1)$$

where ϵ_{eff} is the relative effective dielectric, w_0 is the microstrip line width, and h is the substrate thickness.

With the characteristic impedance $Z_0 = 50 \Omega$, the feeding line width is calculated as $W_{ms} = 1.49$ mm.

Figure 2 presents the simulated $|S_{11}|$ of a single antenna with calculated L_d and W_{ms} values. It can be observed that the antenna operates at a frequency of 4.91 GHz. However, the impedance matching is not satisfactory. As can be seen from this figure, the antenna can broaden its impedance bandwidth thanks to an

extra resonance. To address this issue, the dipole feeding probe position, which is defined as O , will be investigated to optimize the impedance matching as well as the operating bandwidth of n79 frequency band (C-band).

The simulated results presented in Figure 3 indicated that the deep resonance of the single antenna can also be adjusted by changing the feeding position O . This proves that the impedance matching of the single antenna can be optimized by changing the feeding position of L-shaped probe feed.

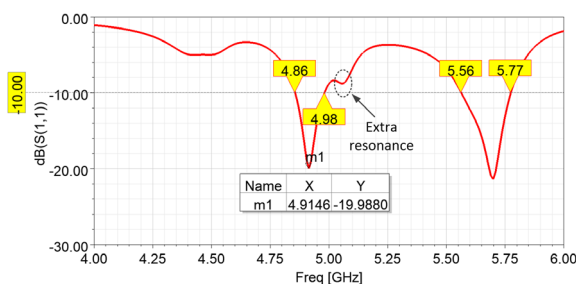


Figure 2. Simulated $|S_{11}|$ of single antenna with initial $L_d = 14.9$ mm.

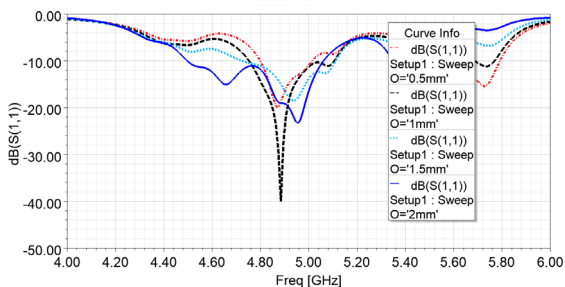
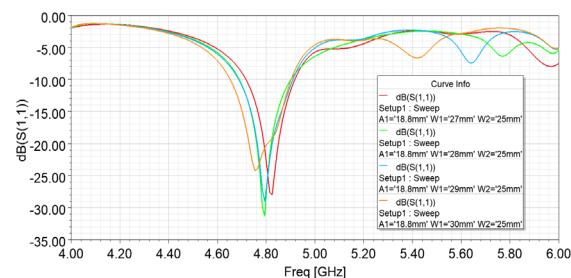
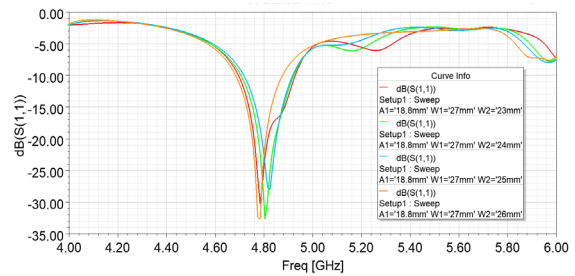


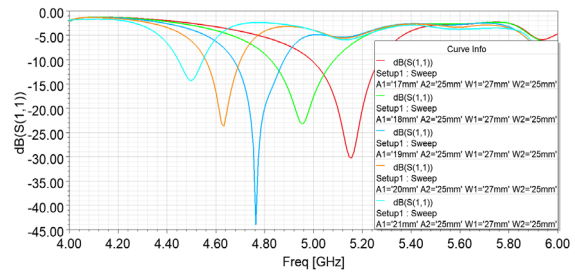
Figure 3. Simulated $|S_{11}|$ of single antenna for different O .



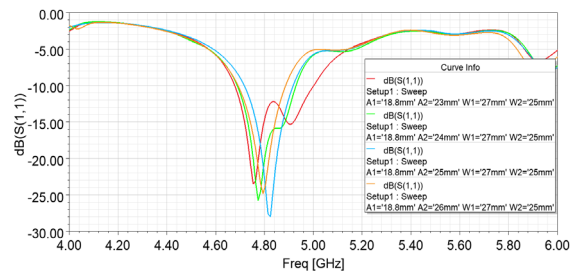
(a)



(b)



(c)



(d)

Figure 4. Simulated $|S_{11}|$ of single antenna for different: (a) W_1 , (b) W_2 , (c) A_1 , (d) A_2 .

Figure 4 presents the $|S_{11}|$ simulated of single ME antenna with different value of W_1 , W_2 , A_1 , and A_2 . As can be observed at Fig. 4(a) and 4(b), the center frequency of single antenna almost did not change when W_1 , W_2 varied. As shown in Fig. 4(c), when A_1 was increased, the center frequency of the single antenna was decreased while this center frequency shifted slightly when A_2 varied as shown in Figure 4(d). This proves that the center frequency of the single antenna can be effectively adjusted by changing the values of A_1 .

The simulated $|S_{11}|$ of final single antenna is shown in Fig. 5. It can be seen that the antenna exhibits a -10 dB impedance bandwidth ranging

from 4.50 GHz to 5.06 GHz. Fig. 6 presents the simulated radiation pattern of single antenna in x-z and y-z plane. It is observed that the antenna yielded an omnidirectional in both planes with a peak gain of 3.1 dBi.

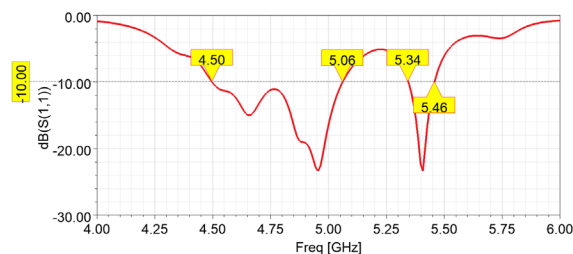


Figure 5. Simulated $|S_{11}|$ of single antenna.

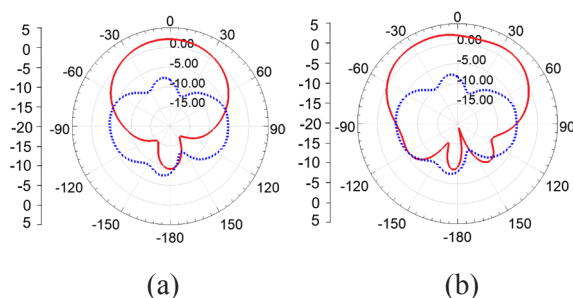


Figure 6. Simulated radiation pattern of the single dipole at 4.9 GHz in (a) x-z plane, (b) y-z plane.

The final design parameters of the single antenna were as follows: $W_{GND}=96\text{ mm}$, $W_1=27\text{ mm}$, $W_2=25\text{ mm}$, $A_1=18.8\text{ mm}$, $A_2=23\text{ mm}$, $W_{ms}=1.49\text{ mm}$, $L_p=15\text{ mm}$, $W_p=5\text{ mm}$, $g_1=1.0\text{ mm}$, $g_2=7.0\text{ mm}$, $O=2\text{ mm}$, and $p=4.5\text{ mm}$.

3. ARRAY ANTENNA DESIGN

In this section, we first analyze the performance of the array by utilizing an in-phase equal-magnitude feeding network. Subsequently, we implement an in-phase tapering-magnitude feeding network to reduce the side-lobe levels of a 1×8 element array antenna. Both feeding networks are designed on FR4 epoxy material with a thickness of 0.8 mm.

3.1. Conventional feeding network

A conventional feeding network consisting of multiple equal T-junction power dividers is employed to achieve in-phase, equal-magnitude excitation for the 1×8 element array antenna. As illustrated in Figure 7, the 1×8 array exhibits

a directional radiation pattern in the y-z plane, achieving a peak gain of 13.8 dBi. However, as shown in Figure 8, the array exhibits a relatively high side-lobe level of -12.9 dB .

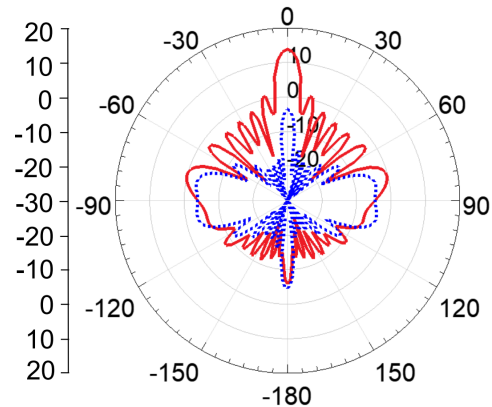


Figure 7. Simulated radiation pattern of 1×8 elements array at 4.9 GHz with an in-phase equal-magnitude feeding (solid line: co-pol, dashed line: cross-pol).

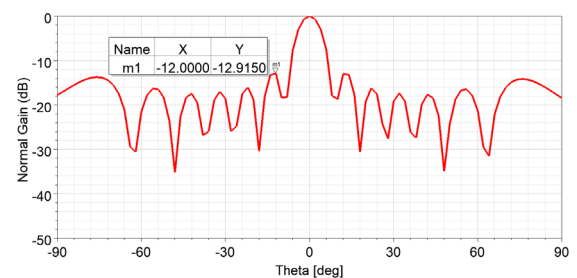


Figure 8. Simulated normal gain of the 1×8 array at 4.9 GHz with an in-phase, equal-magnitude feeding.

3.2. Tapering-magnitude feeding network

In order to reduce the side-lobe, a new magnitude distribution of the excitation, which is type trapezoidal distribution, is designed for the 1×8 elements linear arrays. The feeding network is constructed using a cascade of multiple unequal T-junction power dividers to realize the desired trapezoidal amplitude excitation. The design of the unequal T-junctions is based on impedance transformation principles, in which the characteristic impedances of the microstrip transmission lines are carefully calculated to achieve the desired power division ratios. Quarter-wavelength transmission line transformers are employed for impedance matching between stages. To circumvent fabrication issues associated with extremely

narrow microstrip lines-typically required for high impedance values-each power divider incorporates a $50\ \Omega$ to $25\ \Omega$ transformation section. Detailed feeding network is illustrated in Figure 9. The magnitude for each element is detailed in Table 1.

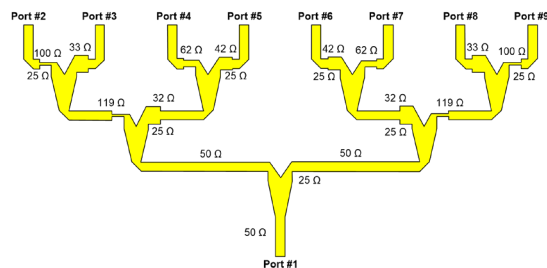


Figure 9. Feeding network of the trapezoidal excitation.

Table 1. Tapering-magnitude excitation for 1×8 ME antenna array.

Element no.	#1	#2	#3	#4	#5	#6	#7	#8
Magnitude	1	3	6	9	9	6	3	1

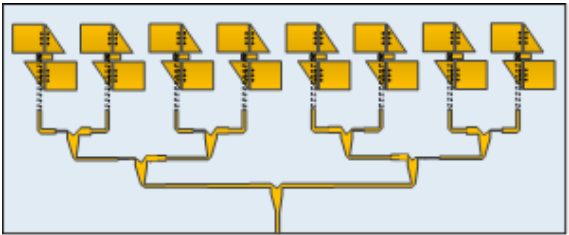


Figure 10. Geometry of 1×8 elements linear array fed by in-phase and tapering-magnitude feeding network.

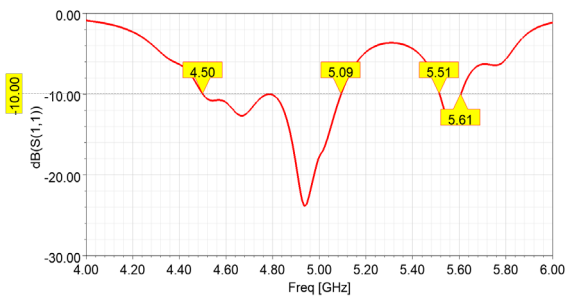


Figure 11. Simulated $|S_{11}|$ of 1×8 elements array.

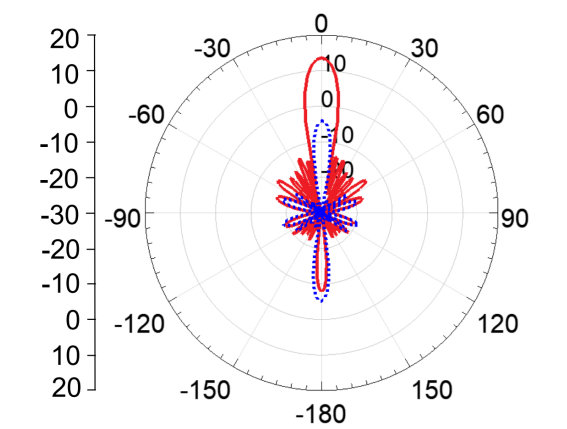


Figure 12. Simulated radiation pattern simulated of 1×8 array at 4.9 GHz (solid line: co-pol, dashed line: cross-pol) with an in-phase trapezoidal feeding.

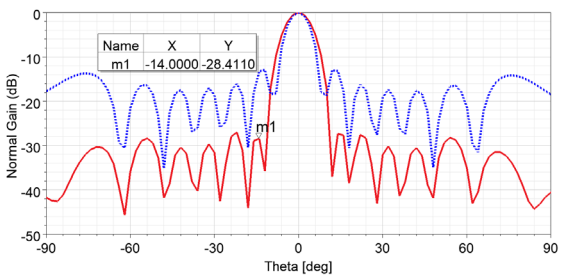


Figure 13. Simulated normal gain of 1×8 elements array at 4.9 GHz with an in-phase trapezoidal feeding.

Figure 10 shows the 1×8 elements array incorporated with in-phase and tapering-magnitude feeding network. The array antenna exhibits a -10 dB bandwidth of 509 MHz, ranging from 4.50 GHz to 5.09 GHz, as illustrated in Figure 11. The simulated radiation performance of the array with the trapezoidal excitation is given in Figure 12. As can be observed that the array yielded a gain of 13.6 dBi and a side-lobe of -28.4 dB. These results indicate that the 1×8 elements linear array with the new excitation yields a lightly lower gain, but a significantly lower side-lobe in comparison with the array fed by in-phase equal-magnitude excitation, as depicted in Figure 13.

4. CONCLUSION

A high-gain, low side-lobe level magneto-electric (ME) array antenna has been developed for base station applications operating in the n79

frequency band (C-band) of the 5G sub-6 GHz communication spectrum. The proposed antenna structure comprises two rectangular and two triangular metallic radiating elements, six grounded vias, an L-shaped feeding probe, a ground plane, and a microstrip line feed. The single-element antenna achieves an impedance bandwidth of 11.4% (ranging from 4.50 GHz to 5.06 GHz) for $|S_{11}| < -10$ dB, along with a peak gain of 3.1 dBi. Subsequently, a linear array consisting of 1×8 elements was designed using a conventional feeding network based on multiple equally split T-junctions. This array configuration achieved a high peak gain of 13.8 dBi but suffered from a relatively high side-lobe level (SLL) of -12.9 dB. To mitigate this issue, a modified excitation scheme employing a trapezoidal amplitude distribution was implemented for the linear ME dipole array. Simulation results indicate that the revised configuration achieves a peak gain of 13 dBi and a significantly reduced side-lobe level, which is below -28 dB.

Acknowledgments

This research is conducted within the framework of science and technology projects at institutional level of Quy Nhon University under the project code T2024.846.17.

REFERENCES

1. T. Liu, W. J. Zhu, , X. J. Hu, , L. H. Ye, D. L. Wu. Dual-wideband dual-polarized magnetoelectric dipole antenna for sub-6 GHz applications, *IEEE Antennas and Wireless Propagation Letters*, **2023**, 22(6), 1396-1400.
2. S. I. Siddiqui, S. Bashir, A. Khan, S. Ghafoor, I. Aziz. A dual-band high-gain beam steering antenna array for 5G sub-6 GHz base station, *Scientific Reports*, **2024**, 14(1), 26517.
3. D. Yang, H. Zhai, C. Guo, C. Ma. A novel differentially fed dual-polarized filtering magneto-electric dipole antenna for 5G base station applications, *IEEE Transactions on Antennas and Propagation*, **2022**, 70(7), 5373-5382.
4. L. Zhao, H. Zhu, H. Zhao, G. Liu, K. Wang, J. Mou, W. Zhang, J. Li. Design of wideband dual-polarized ME dipole antenna with parasitic elements and improved feed structure, *IEEE Antennas and Wireless Propagation Letters*, **2023**, 22(1), 174-178.
5. G. Scalise, E. Arnieri, G. Amendola, M. W. Rousstia, S. Pires, L. Boccia. Dual-polarized magneto-electric dipole for 5G 28 GHz phased array applications, *IEEE Open Journal of Antennas Propagation*, **2023**, 5(1), 164-179.
6. M. Li, K. Luk. Wideband magneto-electric dipole antenna for 60-GHz millimeter-wave communications, *IEEE Transactions on Antennas and Propagation*, **2015**, 63(7), 3276-3279.
7. Y. Li, K. M. Luk. A 60-GHz wideband circularly polarized aperture-coupled magneto-electric dipole antenna array, *IEEE Transactions on Antennas and Propagation*, **2016**, 64(4), 1325-1333.
8. L. Zhao, Y. Li, J. Wang, L. Ge. A study on millimeter-wave magneto-electric dipole phased arrays for 5G dual-band applications, *IEEE Transactions on Antennas and Propagation*, **2022**, 71(3), 2375-2384.
9. D. A. Sehrai, J. Khan, M. Abdullah, M. Asif, M. Alibakhshikenari, B. Virdee, W. A. Shah, S. Khan, M. Ibrar, S. Jan, A. Ullah, F. Falcone. Design of high gain base station antenna array for mm-wave cellular communication systems, *Scientific Reports*, **2023**, 13(1), 4907.
10. L. Zhao, H. Zhu, H. Zhao, G. Liu, K. Wang, J. Mou, W. Zhang, J. Li. Design of wideband dual-polarized ME dipole antenna with parasitic elements and improved feed structure,

IEEE Antennas and Wireless Propagation Letters, **2023**, 22(1), 174-178.

11. D. Yang, H. Zhai, C. Guo, C. Ma. A novel differentially fed dual-polarized filtering magneto-electric dipole antenna for 5G base station applications, *IEEE Transactions on Antennas and Propagation*, **2022**, 70(7), 5373-5382.
12. Z. Song, J. Qi. A novel dual-polarized magnetoelectric dipole antenna and its array for LTE and 5G sub-6 GHz base station applications, *Entropy*, **2023**, 25(2), 274.
13. T. Liu, W. J. Zhu, X. J. Hu, L. H. Ye, D. L. Wu. Dual-wideband dual-polarized magnetoelectric dipole antenna for Sub-6 GHz applications, *IEEE Antennas and Wireless Propagation Letters*, **2023**, 22(6), 1396-1400.
14. B. Feng, K. L. Chung, J. Lai, Q. Zeng. A conformal magneto-electric dipole antenna with wide H-plane and band-notch radiation characteristics for sub-6-GHz 5G base-station, *IEEE Access*, **2019**, 7, 17469-17479.
15. M. Li, K. M. Luk. Analysis of a wideband magneto-electric dipole antenna with low cross polarization and high gain for base station applications, *IEEE Transactions on Antennas and Propagation*, **2018**, 66(1), 67-76.
16. D. M. Pozar. *Microwave engineering: theory and techniques*, John Wiley & Sons, Hoboken, NJ, 2021.



© 2025 by the authors. This Open Access Article is licensed under the Creative Commons Attribution-NonCommercial 4.0 International (CC BY-NC 4.0) license (<https://creativecommons.org/licenses/by-nc/4.0/>).

Tổng hợp màng nhựa sinh học từ cao chiết lá Ngũ trảo dựa trên nền tinh bột Khoai lang và khảo sát hoạt tính kháng khuẩn của sản phẩm

Phạm Khánh Nguyên Huân[#], Lê Hồng Ngọc Bảo[#],
Nguyễn Thị Kiều Tiên, Hà Thị Kim Quy^{*}

Khoa Khoa học Tự nhiên, Trường Đại học Cần Thơ, Việt Nam

Ngày nhận bài: 15/04/2025; Ngày sửa bài: 09/08/2025;
Ngày nhận đăng: 03/09/2025; Ngày xuất bản: 28/10/2025

TÓM TẮT

Ô nhiễm nhựa và an toàn thực phẩm là hai thách thức toàn cầu đòi hỏi các giải pháp bền vững. Nghiên cứu này nhằm phát triển màng nhựa sinh học đa chức năng từ tinh bột khoai lang kết hợp cao chiết lá Ngũ trảo (*Vitex negundo* L.) có hoạt tính kháng khuẩn. Quá trình tối ưu hóa xác định nồng độ thích hợp của chất hóa dẻo glycerol là 10% (w/w), kết hợp với 10% cao chiết tạo ra màng nhựa có đặc tính cơ-lý phù hợp với độ bền kéo đạt 4,26 MPa và khả năng thấm nước thấp. Phân tích FT-IR và SEM xác nhận sự kết hợp thành công của cao chiết vào cấu trúc phân tử của màng, với bề mặt đồng nhất không có vết nứt. Đặc biệt, màng nhựa thể hiện hoạt tính kháng khuẩn đối với cả vi khuẩn Gram dương (*Listeria innocua*) và Gram âm (*Escherichia coli* O157:H7, *Salmonella enterica*) - những tác nhân chính gây ngộ độc thực phẩm. Quan trọng hơn, hoạt tính kháng khuẩn vẫn duy trì sau 3 tháng lưu trữ, chứng minh tính ổn định dài hạn của sản phẩm. Kết quả của nghiên cứu này mở ra triển vọng ứng dụng rộng rãi cho vật liệu bao gói thực phẩm thân thiện với môi trường, có khả năng kéo dài thời gian bảo quản thực phẩm và giảm thiểu nguy cơ nhiễm khuẩn. Màng nhựa sinh học này không chỉ góp phần đưa ra giải pháp cho vấn đề ô nhiễm nhựa mà còn tận dụng được nguồn nông sản sẵn có tại địa phương để sản xuất vật liệu, hướng tới mục tiêu phát triển bền vững.

Từ khóa: Màng nhựa sinh học, Ngũ trảo, chất hóa dẻo glycerol, hoạt tính kháng khuẩn.

^{*}Tác giả liên hệ chính.

Email: htkquy@ctu.edu.vn;

[#]Đồng tác giả đầu tiên.

Synthesis of bioplastic films from *Vitex negundo* L. leaf extract based on sweet potato starch and investigation of antibacterial activity of the products

Pham Khanh Nguyen Huan[#], Le Hong Ngoc Bao[#],
Nguyen Thi Kieu Tien, Ha Thi Kim Quy^{*}

College of Natural Sciences, Can Tho University, Vietnam

Received: 15/04/2025; Revised: 09/08/2025;

Accepted: 03/09/2025; Published: 28/10/2025

ABSTRACT

Plastic pollution and food safety are global challenges requiring sustainable solutions. This study aimed to develop a multifunctional bioplastic film from sweet potato starch combined with *Vitex negundo* L. leaf extract with antimicrobial properties. Optimization processes determined that 10% (w/w) glycerol plasticizer combined with 10% extract produced films with appropriate physicochemical properties, including tensile strength of 4.26 MPa and low water absorption. FT-IR and SEM analyses confirmed the successful incorporation of the extract into the molecular structure of the film, with a homogeneous surface free of cracks. Notably, the bioplastic film exhibited antimicrobial activity against both Gram-positive (*Listeria innocua*) and Gram-negative bacteria (*Escherichia coli* O157:H7, *Salmonella enterica*) - key foodborne pathogens. Importantly, this antimicrobial activity was maintained after three months of storage, demonstrating the long-term stability of the product. The results of this study reveal extensive potential applications for environmentally friendly food packaging materials capable of extending food preservation time while reducing contamination risks. This bioplastic film not only contributes to providing solutions for plastic pollution but also utilizes locally available agricultural resources for material production, supporting sustainable development goals.

Keywords: *Bioplastic films, Vitex negundo L., glycerol as a plasticizer, antibacterial activity.*

1. INTRODUCTION

Plastic pollution has emerged as one of the most serious environmental challenges of the 21st century. Since the 1950s, approximately 8.3 billion tons of plastic have been manufactured, with around 60% now considered waste in landfills or the natural environment.¹ Annually, an estimated 8 million tons of plastic enter the

oceans, resulting in severe repercussions for marine ecosystems and hazards to human health via the food chain.^{2,3} Microplastics have been detected in many marine organisms, in the air, drinking water, and the food daily consume.⁴⁻⁶ Besides the threat of plastic pollution, an even more urgent issue is the dependence of the plastic industry on non-renewable fossil fuels. Today's

^{*}Corresponding author.

Email: htkquy@ctu.edu.vn,

[#]Co-first authors.

commercial plastic products from petroleum cause considerable greenhouse gas emissions.⁷ The long-term sustainability of the plastic industry requires alternatives from renewable and environmentally friendly resources.

Bioplastic films from natural raw materials have been attracting increasing research interest as a potential solution for environmental issues related to traditional plastics.⁸ Bioplastics originate from renewable materials and are biodegradable, reducing ecological impact after disposal.⁹ Among natural raw materials, plant polysaccharides, especially starch, stand out as an attractive choice for bioplastic production due to their abundance, low cost, and complete biodegradability.¹⁰

Sweet potato (*Ipomoea batatas* L.) starch is a promising polysaccharide source for bioplastic production. Sweet potatoes are widely grown in many countries, including Vietnam. This crop has a high starch content and is suitable for producing biopolymer materials. Sweet potato starch has an appropriate amylose/amylopectin ratio, affecting the physicochemical properties of the bioplastic films created from it.¹¹ Sweet potato starch has good viscosity and gelation properties, suitable for producing films with good mechanical strength.¹² However, a significant limitation of starch-based materials is their sensitivity to water and limited mechanical strength compared to synthetic plastics.¹³ To improve these limited characteristics of starch-based bioplastic films, many studies have focused on using various plasticizers and additives such as glycerol, sorbitol, and chitosan^{14–16} to enhance the properties of bioplastic films. Another promising research direction is the development of bioplastic films with antimicrobial properties for food packaging applications. Several studies have proposed the potential of plant extracts or essential oils in creating antimicrobial bioplastic films.¹⁷ However, using plant extracts with biological activity to enhance the antimicrobial capability of starch-based plastic films remains an interesting field for further research.

Vitex negundo L. is a medicinal plant widely used in traditional medicine in many Asian countries. *V. negundo* leaves contain many biologically active compounds, such as flavonoids, terpenoids, glycosides, and alkaloids.^{18–20} Several studies have demonstrated that *V. negundo* leaf extract has good antimicrobial (such as *Escherichia coli*, *Staphylococcus aureus*, *Klebsiella pneumoniae*, *Vibrio cholerae*, *Vibrio parahaemolyticus*, *Vibrio mimicus*, *Shigella* spp, and *Aeromonas* spp) and antioxidant capabilities.^{21–23} Although there have been many studies on the biological activity of *V. negundo* leaves and the potential of sweet potato starch in bioplastic film production, the combination of these two materials to create bioplastic films with antimicrobial potential remains an area that needs to be researched to develop environmentally friendly materials useful for food packaging applications.

2. MATERIALS AND METHODS

2.1. Preparation and extraction of *V. negundo*

V. negundo leaves were collected in Can Tho City and identified by Dr. Pham Ha Thanh Tung (Phenikaa University). After removing damaged leaves, the leaves were washed, dried, and ground into powder to obtain the raw material. The powder was placed in a cloth bag and soaked in 96° EtOH solvent for 72 hours. Then, the extract was dried with Na₂SO₄ and filtered through filter paper. The soaking extraction process was repeated two more times. The extract was concentrated by rotary evaporation to recover the solvent, obtaining the EtOH extract.

2.2. Extraction of sweet potato starch

White sweet potatoes purchased from markets in Can Tho City were washed and ground into a paste. Then, distilled water was added to the sweet potato mixture, and a filter cloth was used to squeeze out the solution containing starch. The filtered starch solution was held at room temperature to allow the starch sediment to settle. After about 5 hours, the water was removed, obtaining the starch in paste form. The

starch was washed 3 times with water, dried at 40°C, and then finely ground.²⁴

2.3. Preparation of bioplastic films

5.0 g of sweet potato starch was dispersed in 50 mL of distilled water, adding glycerol or sorbitol at different concentrations (10%, 20%, or 30% w/w). The mixture was stirred using a heated magnetic stirrer (Phoenix Instrument RSM-01HP) for 25 minutes at 75°C until a homogeneous mixture was obtained. It was then blended with *V. negundo* leaf extract at different concentrations (0%, 10%, 20%, or 30% w/w). This mixture was poured into the molds and dried at room temperature for 2 days, then dried at 50°C for 10 minutes. The resulting plastic films were removed from the molds and stored in sealed containers with desiccant for subsequent experiments.

2.4. Investigation of sensory, physical, and chemical properties of bioplastic films

2.4.1. Sensory evaluation and thickness measurement

A sensory evaluation was conducted regarding the feeling upon contact and the texture of the plastic film samples at different extract concentrations.

The thickness of plastic film samples without extract and with extract at different concentrations was measured using an electronic thickness gauge with 0.01 mm accuracy. Thickness measurements were taken at 5 random locations on the plastic film sample.

2.4.2. Water absorption measurement

Water absorption measurement was performed according to the ASTM D570 standard with modifications to suit the current sample condition better. Plastic film samples at different extract concentrations (0, 5, 10, 20, or 30% w/w) were cut into 2 cm × 2 cm samples, then weighed and recorded the dry weight before testing. Those samples were immersed in 20 mL of distilled water. At specific time points, the samples were removed and wiped with a dry cloth before the wet weights were recorded.²⁴ The experiment was repeated 3 times. The percentage of water absorbed was calculated according to the formula:

$$\% \text{ Water absorption} = (\text{Wet weight} - \text{Dry weight}) / \text{Dry weight} \times 100.$$

2.4.3. Tensile strength measurement

The tensile strength of the plastic film was determined based on the ASTM D882 standard. The results were expressed in N.

2.4.4. Fourier-transform infrared spectroscopy (FT-IR) analysis

The plastic films were analyzed by FT-IR spectroscopy using an FT/IR-4600typeA instrument, with spectra recorded from 4000 cm⁻¹ to 400 cm⁻¹ and 16 scans. The two types of plastic films were measured as film sheets, and the extract was measured as a KBr pellet.

2.4.5. SEM Scanning electron microscopy (SEM) analysis

Scanning electron microscopy (JCM 7000, JOEL, Japan) was used to analyze the surface morphology of the bioplastic films.

2.5. Antibacterial activity testing

2.5.1. Evaluation of the antibacterial activity of *V. negundo* leaf EtOH extract

The agar well diffusion method is a simple method widely used to evaluate antibacterial activity. Similar to the procedure used in the disk diffusion method, the surface of the agar plate is spread with bacterial suspension across the entire agar surface. After the agar plate has been inoculated, wells with diameters of 8 mm are created using a sterile tool, and an amount of antimicrobial agent or extract at the test concentration is introduced into the wells. Then, the agar plates are incubated under appropriate conditions depending on the test microorganism. The antimicrobial agent diffuses in the agar medium and inhibits the growth of the test microbial strain.

Preparation: TSB (tryptic soy broth) or TSA (tryptic soy agar) bacterial culture media were prepared. The TSB liquid medium was prepared in deionized water at a ratio of 3% (w/v), and the TSA medium was supplemented with 1.5% (w/v) agar. Sterilization was performed at 121°C for 20 minutes.

Procedure: Bacterial strains were cultured for 24 hours in TSB medium. Then, the bacterial suspension was transferred to a new liquid medium tube to achieve a bacterial density of approximately 0.5 McFarland Units. 100 µL of bacterial suspension was pipetted onto TSA agar plates and spread evenly. After the plates were dried, wells of 8 mm diameter were created, and 100 µL of extract dissolved in 20% DMSO was added to each well. Similarly, 100 µL of tetracycline 0.1 mg/mL (positive control) or 20% DMSO (negative control) was added.

Results were read after 24 hours of incubation at 32°C, measuring the inhibition zones and taking the average value of 3 replicates. The larger the diameter of the inhibition zone, the stronger the antibacterial activity, and vice versa. The diameter of the inhibition zone (DIZ) was calculated according to the formula:

$$\text{DIZ (mm)} = D - d$$

Where: D: diameter of the bacterial inhibition zone, including the diameter of the well (mm); d: diameter of the well (d = 8 mm).

2.5.2. Investigation of antibacterial activity of bioplastic films

Media preparation: Soft agar consisting of 3% TSB (w/v) and 0.5% agar (w/v), sterilized at 121°C for 20 minutes. After sterilization, the soft agar was maintained at a stable temperature of 50°C before use.

Procedure: 100 µL of bacterial suspension at a concentration of 0.5 McFarland Units ($\sim 1.5 \times 10^7$ CFU/mL) was added to 4 mL of soft agar solution, mixed well, and immediately poured

onto a hard agar plate, then waited for the plate to solidify and dry. The pre-prepared plastic film samples with a diameter of 10 mm were placed on the surface of the soft agar. The negative control was plastic film without extract (sample diameter 10 mm) and plasticized film. The agar plates were incubated for 24 hours at 32°C and then observed for the antibacterial ability of the plastic films.

3. RESULTS AND DISCUSSION

3.1. Extraction of *V. negundo* leaf extract and sweet potato starch

From 1.0 kg of dry *V. negundo* leaf powder, approximately 200 g of extract was obtained using 96° EtOH solvent maceration, yielding an extraction efficiency of approximately 20%. To utilize readily available agricultural resources, we selected sweet potatoes from local markets as our starch source in this study. From 10 kg of fresh sweet potatoes, after extraction, approximately 1.8 kg of sweet potato starch was obtained. The extraction yield was approximately 18%.

3.2. Optimization of bioplastic film synthesis conditions

Natural polymers, such as polysaccharides, can form bioplastic films. However, bioplastic films synthesized from starch are typically brittle and easily broken. Hence, plasticizers need to be added to the film structure to improve the mechanical properties of the bioplastic films. Glycerol and sorbitol at different concentrations were selected as plasticizers in this study to investigate the appropriate supplementation concentration on the starch base as a foundation for subsequent studies, combined with *V. negundo* leaf extract.

Table 1. Sensory evaluation and thickness of bioplastic films containing glycerol and sorbitol plasticizers.

Plasticizer	Sample Concentration (% w/w)	Thickness (mm)	Color	Texture	Sensory Evaluation
Glycerol	5	0.21 ± 0.06 ^{bc}	White	Brittle	Not good
	10	0.19 ± 0.03 ^c		Flexible	Good
	15	0.28 ± 0.04 ^{ab}		Flexible	Good
	20	0.26 ± 0.03 ^{abc}		Moist, soft	Not good
	30	0.33 ± 0.06 ^a		Soft, sticky	Not good
Sorbitol	5	0.24 ± 0.02 ^A	White	Brittle	Not good
	10	0.22 ± 0.05 ^A		Flexible	Good
	15	0.25 ± 0.01 ^A		Flexible	Good
	20	0.23 ± 0.04 ^A		Moist, soft	Not good

Values not sharing the same letter are significantly different ($p < 0.05$).

Based on the results shown in Table 1, the film thickness ranged from 0.19 mm to 0.33 mm for glycerol plasticizer and from 0.22 mm to 0.25 mm for sorbitol plasticizer. These results demonstrate that for glycerol plasticizer, as the concentration of plasticizer increased, the thickness of the film also increased. Because glycerol can form hydrogen bonds with water molecules, increasing the volume of the plastic film, the sample thickness increases accordingly when the glycerol concentration increases. Meanwhile, sorbitol primarily forms hydrogen bonds with polymer chains, forming fewer bonds with water than glycerol, so plastic films using sorbitol plasticizers remain of similar thickness while increasing sorbitol concentration.²⁵ Besides the hydrogen bonding interactions, the observed thickness variations can be attributed to differences in solution viscosity and homogeneity between plasticizers. Glycerol exhibits significantly higher viscosity (~1400 cP) compared to sorbitol (200-300 cP) at room temperature, resulting in slower solvent evaporation and reduced solution spreading during film-making, which leads to thicker films with greater thickness variation. Therefore, glycerol-plasticized films demonstrate significant thickness differences between concentrations ($p < 0.05$), whereas sorbitol films maintain consistent thickness across all concentrations tested.

For bioplastic films containing 5% glycerol or 5% sorbitol, the products formed were brittle and easily broken when subjected to external forces. Films containing 20% and 30% glycerol or 20% sorbitol had soft textures and were easily torn. Therefore, films at those concentrations did not meet the requirements for strength and flexibility for subsequent further studies.

Films containing 10% and 15% glycerol or sorbitol could be bent and had appropriate flexibility; additionally, in terms of sensory evaluation, the film surface maintained dry characteristics upon contact. Therefore, films containing glycerol or sorbitol at these concentrations met the requirements in terms of sensory evaluation. However, when considering the economic aspects of the product, 10% glycerol or sorbitol concentration was determined to be more reasonable.

We propose that films with 10% glycerol or sorbitol are the most suitable for combining with different concentrations of *V. negundo* leaf extract (Table 1). As seen in Table 2, when the extract is incorporated into the film, the thickness of the product increases. In addition, a clear difference can be observed between films using glycerol or sorbitol as plasticizers when the extract is added. The film thickness changes significantly when the extract concentration increases for glycerol films. However, the difference is insignificant when combining extract into films containing sorbitol, although the film thickness increases, as demonstrated by the statistical results in Table 2.

Based on the sensory evaluations of texture, strength, and flexibility of the different types of films, it can be seen that films containing 5% and 10% extract with two kinds of plasticizers (glycerol or sorbitol) meet the sensory requirements as they have flexibility when bent and folded, and dry surface characteristics.

Films containing higher concentrations of the extract showed unacceptable results during sensory evaluation due to either too soft texture or excessively sticky surface characteristics.

Table 2. Sensory Evaluation and thickness of bioplastic films containing 10% glycerol and 10% sorbitol combined with *V. negundo* leaf extract.

Sample		Thickness (mm)	Color	Texture	Sensory Evaluation
Plasticizer	Concentration (%, w/w)				
10% Glycerol	5	0.16 ± 0.01 ^b	Green	Flexible	Good
	10	0.184 ± 0.04 ^b	Green	Flexible	Good
	20	0.23 ± 0.03 ^b	Green	Moist, soft	Not good
	30	0.36 ± 0.11 ^a	Dark green	Moist, soft	Not good
10% Sorbitol	5	0.22 ± 0.02 ^A	Green	Slightly flexible	Good
	10	0.24 ± 0.04 ^A	Green	Slightly flexible	Good
	20	0.26 ± 0.01 ^A	Green	Moist, soft	Not good
	30	0.23 ± 0.02 ^A	Dark green	Moist, soft	Not good

Values not sharing the same letter are significantly different ($p < 0.05$).

3.3. Results of physical-chemical properties investigation of bioplastic films

3.3.1. Water absorption

Films containing 5% and 10% extract demonstrated the lowest water absorption among all tested bioplastic films, while those without extract (containing only 10% glycerol) exhibited the highest water absorption (Figure 1). The water absorption manner followed a consistent pattern: all films absorbed water rapidly during the first 5 minutes of immersion, followed by minimal additional absorption from minute 10 to 60, as evidenced by the nearly horizontal curve relative to the x-axis, indicating that the films reached water saturation. Despite 60 minutes of water immersion, all films maintained their structural integrity, remaining firm and resistant to tearing or pulling forces.

Similar to glycerol-containing films, the sorbitol-containing films exhibited strong water absorption during the first 5 minutes, followed by minimal absorption from minutes 10 to 60, eventually reaching saturation (Figure 2). After 60 minutes of water immersion, all films maintained their structural integrity, remaining firm and resistant to tearing or pulling forces. Notably, glycerol-containing films showed clear differences in water absorption percentages among formulations, whereas sorbitol-containing films demonstrated more uniform water absorption capabilities across different formulations.

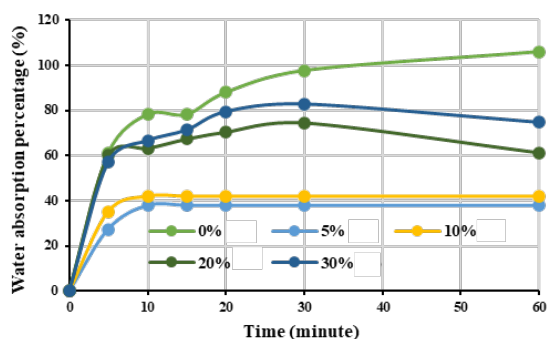


Figure 1. Water absorption percentage of bioplastic films containing 10% glycerol with extract additions from 0% to 30%.

Taken together, bioplastic films using glycerol plasticizer supplemented with 5% or 10% extract had the lowest water absorption percentages, indicating the potential application of these products in food preservation.

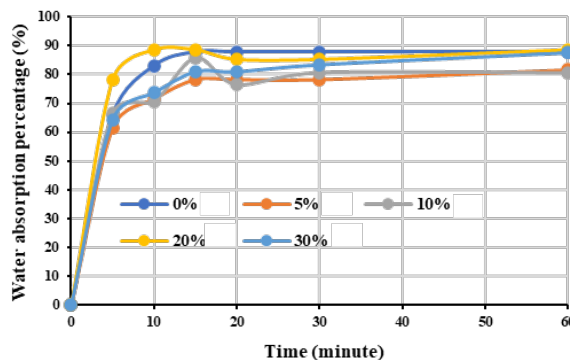


Figure 2. Water absorption percentage of bioplastic films containing 10% sorbitol with extract additions from 0% to 30%.

3.3.2. Tensile strength

Film samples containing 10% glycerol or sorbitol with 10% extract addition were tested for tensile strength using a digital testing device according to the modified ASTM D882 standard. After two months of sample storage, sorbitol-containing films exhibited brittle and easily breakable, so we did not proceed with tensile strength measurements for these samples. At the same time of sample storage, glycerol-containing films still maintained flexibility and elasticity, so tensile strength testing was performed. The results showed that the tensile strength of this film was 4.26 MPa. Compared with results from previous studies, bioplastic films from dialdehyde starch had tensile strengths in the range of 1.63 MPa to 3.06 MPa²⁶, and bioplastic films synthesized from sweet potato starch combined with glycerol plasticizer had a tensile strength of 2.57 MPa¹⁵. It can be seen that the films synthesized in this study have better tensile strength than the compared films.

Based on the tensile strength measurement results, we propose further studies only for the most suitable bioplastic film containing 10% glycerol and 10% extract.

3.3.3. Fourier-Transform Infrared Spectroscopy (FT-IR) analysis

Figure 3 shows the FT-IR spectra of three samples: the EtOH extract, the bioplastic film containing 10% glycerol, and the bioplastic film containing 10% glycerol supplemented with 10% extract. All three spectra display corresponding features. In the wavenumber range of 3500 cm⁻¹ to 3000 cm⁻¹, the signals are attributed to hydroxyl (OH)

groups. Vibrations from 3000 cm^{-1} to 2800 cm^{-1} are assigned to symmetric and asymmetric stretching vibrations of C-H in the CH_2 or CH_3 groups (sp^3 hybridization). Besides, the addition of EtOH extract, which contains many secondary compounds such as polyphenolics, flavonoids, terpenoids and alkaloids, may have brought about small contributions from aromatic C-H vibrations ($\sim 3000\text{ cm}^{-1}$) or additional aliphatic C-H groups from these compounds, causing a slight shift in the spectral peak of the extract-containing bioplastic film. Wavenumbers in 1400 cm^{-1} to 1300 cm^{-1} show signals of asymmetric C-H vibrations in CH_2 and CH_3 groups, and C-O-H bending vibrations. However, notable differences can be observed among the three spectra. Specifically, the FT-IR spectra of the extract and the bioplastic film containing 10%

glycerol supplemented with 10% extract show signals in the range of 1850 cm^{-1} to 1650 cm^{-1} , accounting for the exist of C=O groups and potential C=C stretching from aromatic structures of secondary compounds, whereas the spectrum of the bioplastic film containing only 10% glycerol does not exhibit signals in this region. The differences are likely due to the starch-based film without extract lacking C=O functional groups. At wavenumbers 1300 cm^{-1} to 1000 cm^{-1} , strong absorption signals appear, attributed to C-O groups from starch in both types of films, while the extract shows only weak vibrations in this range. Finally, at wavenumbers 700 cm^{-1} to 900 cm^{-1} , characteristic vibrations of C-O-C groups from starch are observed in both types of films, while the extract does not clearly exhibit vibrations in this range.¹²

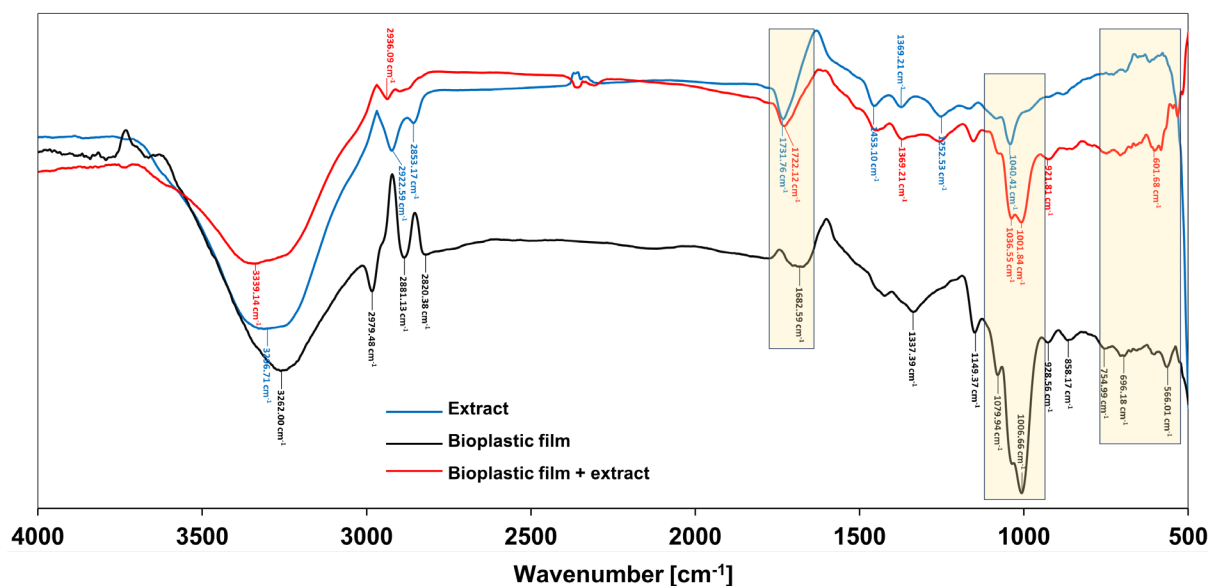


Figure 3. FTIR spectra of *V. negundo* leaf EtOH extract (blue), bioplastic film containing 10% glycerol (black), and bioplastic film containing 10% glycerol with 10% extract addition (red).

3.3.4. Surface morphology analysis by Scanning Electron Microscopy (SEM)

Observation of the SEM images (Figure 4) shows that the surface of the glycerol-plasticized film remains intact when the extract is added, displaying no evidence of tears, fractures, or discontinuities on the film surface.

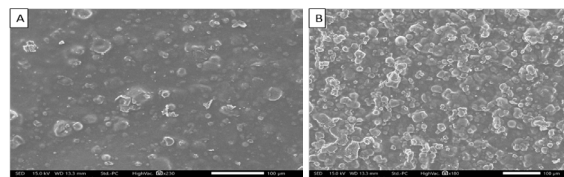


Figure 4. SEM images of bioplastic film containing 10% glycerol (A) at 230 $\times$ magnification and bioplastic film containing 10% glycerol with 10% extract (B) at 180 $\times$ magnification.

3.4. Antibacterial activity results

3.4.1. Antibacterial activity of the extract

Results in Table 3 and Figure 5 demonstrate that *V. negundo* leaf extract at an initial test concentration of 40 mg/mL showed good antibacterial activity against Gram-negative bacteria (*E. coli* O157:H7, *S. enterica* serotype Enteritidis, and *Proteus* sp.). When the extract concentration was increased to 80 mg/mL, the extract exhibited moderate antibacterial activity against other Gram-positive bacteria like *L. innocua* and Gram-negative bacteria (*Campylobacter* sp. and *Citrobacter freundii*). The extract showed weak or no antibacterial activity for some bacterial strains such as *S. aureus*, *S. enterica* serotype Typhimurium, and *Shigella* sp.. The negative control (20% DMSO)

did not exhibit antibacterial activity, while the positive control (tetracycline at 0.1 mg/mL) showed antibacterial activity against almost all bacterial strains used in this study.

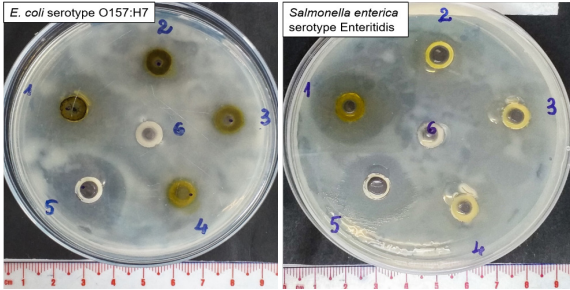


Figure 5. Agar well diffusion results of *E. coli* serotype O157:H7 and *S. enterica* serotype Enteritidis at different concentrations of *V. negundo* leaf extract: (1) 40 mg/mL, (2) 20 mg/mL, (3) 10 mg/mL, (4) 5.0 mg/mL, (5) positive control (tetracycline 0.1 mg/mL), (6) negative control.

Table 3. Antibacterial activity assessment of *V. negundo* leaf extract.

Gram	Bacteria	Inhibition zone diameter (mm)					Tetracycline (0.1 mg/mL)
		Extract (mg/mL)					
		80	40	20	10	5	
(+)	<i>Listeria innocua</i>	6.33 ± 3.51 ^a	4.33 ± 1.16 ^{ab}	3.00 ± 0.00 ^{ab}	2.33 ± 0.58 ^{ab}	NT	6.67 ± 1.53 ^a
	<i>Staphylococcus aureus</i>	+/-	—	—	—	NT	+/-
(-)	<i>Campylobacter</i> sp.	7.00 ±1.41 ^a	4.00 ± 0.00 ^b	2.50 ± 0.71 ^{bc}	0.50 ± 0.71 ^c	NT	6.52 ±1.03 ^a
	<i>Citrobacter freundii</i>	4.33 ± 0.58 ^b	0.67 ± 1.15 ^c	0.33 ± 0.58 ^c	0.33 ± 0.58 ^c	NT	22.33 ± 4.93 ^a
	<i>E. coli</i> serotype O157:H7	NT	22.33 ± 3.21 ^a	13.67 ± 5.13 ^b	3.00 ± 1.73 ^c	1.33 ± 0.58 ^c	13.67 ± 2.89 ^b
	<i>Proteus</i> sp.	NT	10.67 ± 1.53 ^a	6.33 ± 1.16 ^b	2.67 ± 0.57 ^c	NT	4.33 ± 2.01 ^{bc}
	<i>Salmonella enterica</i> serotype Enteritidis	NT	17.0 ± 1.00 ^a	5.00 ± 2.65 ^b	3.33 ± 1.53 ^c	3.00 ± 1.00 ^c	13.67 ± 1.16 ^a
	<i>Salmonella enterica</i> serotype Typhimurium	+/-	—	—	—	NT	15.85 ± 1.69
	<i>Shigella</i> sp.	+/-	—	—	—	NT	20.53 ± 2.74

Note: (-) No inhibition zone observed, (+/-) Weak antimicrobial activity or no clear inhibition zone observed, (NT) concentration not tested for activity. In the row, values not sharing the same letter are significantly different ($p < 0.05$).

3.4.2. Antibacterial activity of bioplastic films

Results in Table 4 show that bioplastic film containing 10% extract demonstrated antibacterial

activity against all three test bacterial strains (*L. innocua*, *E. coli* serotype O157:H7, and *S. enterica* serotype Enteritidis) as evidenced by

reduced bacterial growth on the agar surface, particularly exhibiting good antibacterial activity against *L. innocua* and *S. enterica* serotype Enteritidis. Films containing 5% extract showed good antibacterial results against *S. enterica* serotype Enteritidis while demonstrating moderate antibacterial activity against the other

two bacterial strains.

Film samples stored for 3 months before testing still maintained antibacterial activity against the test bacterial strains. Negative controls (films without extract and plasticized films) showed no antibacterial activity against all four test bacterial strains.

Table 4. Antibacterial activity assessment of bioplastic films containing extract.

Bacteria	Bioplastic films containing extract			Films after 3 months of storage		Plasticized film
	0%	5%	10%	0%	10%	
<i>L. innocua</i>	–	+	++	–	+	–
<i>E. coli</i> serotype O157:H7	–	+/-	+	–	+/-	–
<i>S. enterica</i> serotype Enteritidis	–	++	++	–	+	–

Note: (++) Good antimicrobial activity, (+) Fairly good antimicrobial activity, (+/-) Weak antimicrobial activity, (-) No antimicrobial activity.

Bioplastic films from sweet potato starch combined with *V. negundo* leaf extract have demonstrated significant potential in simultaneously addressing two issues: plastic pollution and food preservation. FT-IR analysis results indicate effective incorporation of the extract into the film structure through the appearance of characteristic signals for C=O groups (1850 cm⁻¹ to 1650 cm⁻¹) in the extract-containing film, while these signals are absent in the starch-only film. These results confirm that the extract is not merely physically dispersed but also chemically interacts with the starch matrix, explaining the stability and ability to maintain antibacterial activity after prolonged storage.²⁷ SEM images show homogeneous film surfaces without cracks when the extract is added, ensuring the structural integrity necessary for packaging applications.

Regarding antibacterial activity, films containing the extract demonstrated effective inhibition against both *L. innocua* (Gram-positive) and *E. coli* O157:H7 and *S. enterica* (Gram-negative). These three bacteria are dangerous foodborne pathogens, particularly *E. coli* O157:H7 and *S. enterica*, which are common causes of severe food poisoning.²⁸ The antibacterial effect may be related to flavonoids, terpenoids, and alkaloids in *V. negundo* leaves, allowing the film to gradually release these compounds, creating a protective barrier for food.

The low water absorption capacity of the extract-containing films (as demonstrated in previous experiments) is an important property that helps overcome the major limitation of traditional starch-based bioplastics. The reduced water uptake can be explained by polyphenolic and terpenoid compounds in the extract forming bonds with hydroxyl groups in starch, limiting their interaction with water.²⁹

The potential applications of antimicrobial bioplastic films from this research could extend beyond food packaging to other fields such as agriculture (seed coating, fertilizer coating) and healthcare (antimicrobial bandages). Youssef and El-Sayed (2018) reported that antimicrobial packaging materials can extend the shelf life of fresh food by 25% to 50%, reducing food waste and enhancing food safety.³⁰ However, for commercialization, further research is needed on large-scale production, durability under real conditions, as well as biological safety assessment when in direct contact with food.³¹

3. CONCLUSION

The research has successfully synthesized biodegradable polymeric films from sweet potato starch incorporated with *V. negundo* leaf extract, exhibiting prominent antimicrobial properties. The biopolymeric matrix containing 10% glycerol and 10% *V. negundo* leaf extract demonstrated an optimal equilibrium between

mechanical characteristics (tensile strength of 4.26 MPa) and aqueous stability. Fourier Transform Infrared Spectroscopy (FT-IR) and Scanning Electron Microscopy (SEM) analyses confirmed the successful molecular integration of starch, glycerol, and phytoextract within the film's supramolecular structure. Notably, the biopolymeric film exhibited efficacious antimicrobial activity against both Gram-positive (*L. innocua*) and Gram-negative (*E. coli* O157, *S. enterica*) bacterial strains - common foodborne pathogens associated with food deterioration and foodborne illness. Significantly, this antimicrobial efficacy persisted after a three-month storage period, exhibiting the long-term stability of the bioactive compounds within the polymeric matrix.

The experimental outcomes elucidate the potential applications of these antimicrobial biopolymeric films in environmentally sustainable food packaging solutions, simultaneously addressing the critical issue of plastic pollution. Subsequent investigations should focus on practical food preservation trials, elucidation of the antimicrobial mechanisms at the molecular level, and process optimization for commercial-scale implementation.

REFERENCES

1. R. Geyer, J. R. Jambeck, K. L. Law. Production, use, and fate of all plastics ever made, *Science Advances*, **2017**, 3, e1700782.
2. J. R. Jambeck, R. Geyer, C. Wilcox, T. R. Siegler, M. Perryman, A. Andrady, R. Narayan, K. L. Law. Plastic waste inputs from land into the ocean, *Science*, **2015**, 347, 768-771.
3. L. M. Ziccardi, A. Edgington, K. Hentz, K. J. Kulacki, S. K. Driscoll. Microplastics as vectors for bioaccumulation of hydrophobic organic chemicals in the marine environment: A state-of-the-science review, *Environmental Toxicology and Chemistry*, **2016**, 35, 1667-1676.
4. N. Von Moos, P. B. Holm, A. Köhler. Uptake and effects of microplastics on cells and tissue of the blue mussel *Mytilus edulis* L. after an experimental exposure, *Environmental Science & Technology*, **2012**, 46, 11327-11335.
5. P. Marsden, A. A. Koelmans, J. B. Lacombe, T. Gouin, L. D'anglada, D. Cunliffe, P. Jarvis, J. Fawell, J. D. France. *Microplastics in drinking water*, World Health Organization, 2019.
6. A. I. Catarino, V. Macchia, W. G. Sanderson, R. C. Thompson, T. B. Henry. Low levels of microplastics (MP) in wild mussels indicate that MP ingestion by humans is minimal compared to exposure via household fibres fallout during a meal, *Environmental Pollution*, **2018**, 237, 675-684.
7. R. Verma, K. S. Vinoda, M. Papireddy, A. N. S. Gowda. Toxic pollutants from plastic waste-a review, *Procedia Environmental Sciences*, **2016**, 35, 701-708.
8. Y. Zhu, C. Romain, C. K. Williams. Sustainable polymers from renewable resources, *Nature*, **2016**, 540, 354-362.
9. H. Karan, C. Funk, M. Grabert, M. Oey, B. Hankamer. Green bioplastics as part of a circular bioeconomy, *Trends in Plant Science*, **2019**, 24, 237-249.
10. U. Qasim, A. I. Osman, A. H. Al-Muhtaseb, C. Farrell, M. A. Abri, M. Ali, D. V. N. Vo, F. Jamil, D. W. Rooney. Renewable cellulosic nanocomposites for food packaging to avoid fossil fuel plastic pollution: a review, *Environmental Chemistry Letters*, **2021**, 19, 613-641.
11. W. Zhou, J. Yang, Y. Hong, G. Liu, J. Zheng, Z. Gu, P. Zhang. Impact of amylose content on starch physicochemical properties in transgenic sweet potato, *Carbohydrate Polymers*, **2015**, 122, 417-427.
12. A. H. D. Abdullah, S. Chalimah, I. Primadona, M. H. G. Hanantyo. *Physical and chemical properties of corn, cassava, and potato starches*, IOP Conference Series: Earth and Environmental Science, IOP Publishing, Jakarta, Indonesia, 2017.
13. R. Thakur, P. Pristijono, C. J. Scarlett, M. Bowyer, S. P. Singh, Q. V. Vuong. Starch-based films: major factors affecting their properties, *International Journal of Biological Macromolecules*, **2019**, 132, 1079-1089.
14. M. G. A. Vieira, M. A. Da Silva, L. O. Dos Santos, M. M. Beppu. Natural-based plasticizers and biopolymer films: a review, *European Polymer Journal*, **2011**, 47, 254-263.

15. A. H. D. Abdullah, S. Pudjiraharti, M. Karina, O. D. Putri, R. H. Fauziyyah. Fabrication and characterization of sweet potato starch-based bioplastics plasticized with glycerol, *Journal of Biological Sciences*, **2018**, *19*, 57-64.
16. A. P. Dewi, A. Mardhiyana, R. Manfaati, U. Leoangraini. The effect of additional chitosan and cellulose on the performance of bioplastic from manihot glaziovii starch, *Fluida*, **2023**, *16*, 36-42.
17. R. Syafiq, S. M. Sapuan, M. Y. M. Zuhri, R. A. Ilyas, A. Nazrin, S. F. K. Sherwani, A. Khalina. Antimicrobial activities of starch-based biopolymers and biocomposites incorporated with plant essential oils: a review, *Polymers*, **2020**, *12*, 2403.
18. B. S. Gill, R. Mehra, Navgeet, S. Kumar. Vitex negundo and its medicinal value, *Molecular Biology Reports*, **2018**, *45*, 2925-2934.
19. N. Koirala, C. Dhakal, N. N. Munankarmi, S. W. Ali, A. Hameed, N. Martins, J. Sharifi-Rad, M. Imran, A. M. Arif, M. S. Hanif. Vitex negundo Linn.: phytochemical composition, nutritional analysis, and antioxidant and antimicrobial activity, *Cellular and Molecular Biology*, **2020**, *66*, 1-7.
20. N. T. Trung, T. P. Nguyen, V. G. Vo, N.-H. Nguyen, T. H. Duong. Further terpenoids from Vitex negundo L. growing in Vietnam, *Journal of Saudi Chemical Society*, **2021**, *25*, 101298.
21. M. S. Deogade, T. Pandya, K. S. Prasad, K. Kale, N. Tankhiwale. Antimicrobial activity of Vitex negundo Linn.(Nirgundi) leaves extract, *Journal of Research in Traditional Medicine*, **2018**, *2*, 99.
22. M. Kamruzzaman, S. M. N. Bari, S. M. Faruque. In vitro and in vivo bactericidal activity of Vitex negundo leaf extract against diverse multidrug resistant enteric bacterial pathogens, *Asian Pacific Journal of Tropical Medicine*, **2013**, *6*, 352-359.
23. O. P. Tiwari, Y. B. Tripathi. Antioxidant properties of different fractions of Vitex negundo Linn., *Food Chemistry*, **2007**, *100*, 1170-1176.
24. T. P. Duy, N. T. Minh, H. G. Ngoc. Synthesis and investigation of some properties of biodegradable materials from potato starch, *Journal of Science on Natural Resources and Environment*, **2021**, 38-46.
25. S. Faust, J. Foerster, M. Lindner, M. Schmid. Effect of glycerol and sorbitol on the mechanical and barrier properties of films based on pea protein isolate produced by high-moisture extrusion processing, *Polymer Engineering & Science*, **2022**, *62*, 95-102.
26. O. O. Oluwasina, B. P. Akinyele, S. J. Olusegun, O. O. Oluwasina, N. D. S. Mohallem. Evaluation of the effects of additives on the properties of starch-based bioplastic film, *SN Applied Sciences*, **2021**, *3*, 1-12.
27. D. P. Hernandez, C. M. Jaramillo, A. L. Córdoba, S. Goyanes. Edible cassava starch films carrying rosemary antioxidant extracts for potential use as active food packaging, *Food Hydrocolloids*, **2017**, *63*, 488-495.
28. V. Kechichian, C. Ditchfield, P. V. Santos, C. C. Tadini. Natural antimicrobial ingredients incorporated in biodegradable films based on cassava starch, *LWT - Food Science and Technology*, **2010**, *43*, 1088-1094.
29. C. Menzel. Improvement of starch films for food packaging through a three-principle approach: Antioxidants, cross-linking and reinforcement, *Carbohydrate Polymers*, **2020**, *250*, 116828.
30. A. M. Youssef, S. M. E. Sayed. Bionanocomposites materials for food packaging applications: concepts and future outlook, *Carbohydrate Polymers*, **2018**, *193*, 19-27.
31. B. Mishra, J. Panda, A. K. Mishra, P. C. Nath, P. K. Nayak, U. Mahapatra, M. Sharma, H. Chopra, Y. K. Mohanta, K. Sridhar. Recent advances in sustainable biopolymer-based nanocomposites for smart food packaging: a review, *International Journal of Biological Macromolecules*, **2024**, 135583.



© 2025 by the authors. This Open Access Article is licensed under the Creative Commons Attribution-NonCommercial 4.0 International (CC BY-NC 4.0) license (<https://creativecommons.org/licenses/by-nc/4.0/>).

MỤC LỤC

1.	Chế tạo vật liệu siêu hấp thụ dựa trên dẫn xuất chitosan ứng dụng trong nông nghiệp Cao Lưu Ngọc Hạnh, Nguyễn Tường Vy, Huỳnh Liên Hương, Nguyễn Minh Nhựt, Phạm Mai Hương	5
2.	Các bóng bán dẫn GAAFET và vật liệu 2D cho công nghệ dưới 3 nm: hiệu năng, thách thức chế tạo và chiến lược tích Huỳnh Công Tú, Hoàng Anh Phương	25
3.	Tổng hợp vật liệu Co_3O_4 có nguồn gốc từ ZIF-67 và ứng dụng trong biến tính điện cực phát hiện acid ascorbic Ngô Thị Thanh Hiền, Nguyễn Vũ Ngọc Mai	43
4.	Chế tạo hạt nano alginate-chitosan chứa ovalbumin bằng phương pháp phun điện đồng trục để định hướng phân phối protein Cao Lưu Ngọc Hạnh, Nguyễn Tường Vy, Phạm Duy Toàn, Nguyễn Thị Bích Thuyền, Đặng Huỳnh Giao, Lê Trường Thịnh	53
5.	Các mô hình mạng nơ ron hồi quy dùng cho dự báo điện gió Nguyễn Văn Ninh, Nguyễn Xuân Thâm, Thân Văn Thông, Đoàn Thanh Bảo, Lương Ngọc Toàn, Lê Tuấn Hộ	69
6.	Nghiên cứu ảnh hưởng của bánh men thuần chủng đến chất lượng sản phẩm cơm rượu Lê Bích Tuyền, Huỳnh Văn Quốc Cảnh, Trương Thị Tú Trân	81
7.	Một đặc trưng của không gian kiểu Zygmund và áp dụng Thái Thuần Quang, Nguyễn Văn Đại	93
8.	Nghiên cứu tối ưu quá trình gia tốc động cơ không đồng bộ ba pha Đỗ Văn Cần, Thân Văn Thông, Hoàng Văn Hiếu	103
9.	Hàm năng lượng tự do của mô hình Ising với từ trường mạnh Nguyễn Tòng Xuân	117
10.	Thiết kế anten mảng trạm gốc có độ lợi cao, mức búp sóng bên thấp cho hệ thống thông tin 5G dải tần dưới 6 GHz Đặng Thị Từ Mỹ, Hồ Văn Phi, Huỳnh Nguyễn Bảo Phương	129
11.	Tổng hợp màng nhựa sinh học từ cao chiết lá Ngũ trảo dựa trên nền tinh bột Khoai lang và khảo sát hoạt tính kháng khuẩn của sản phẩm Phạm Khánh Nguyên Huân, Lê Hồng Ngọc Bảo, Nguyễn Thị Kiều Tiên, Hà Thị Kim Quy	139

