Waste-to-resource conversion of coconut shells into activated carbon for tetracycline removal

ABSTRACT

The increasing occurrence of antibiotic residues in aquatic environments has raised serious ecological and public health concerns, particularly because they promote antibiotic resistance. In this study, coconut shell waste was converted into activated carbon (CSAC) through a thermal carbonization process and applied as a low-cost and sustainable adsorbent for tetracycline (TC) removal from aqueous solutions. The prepared CSAC was comprehensively characterized by SEM–EDX, XRD, FTIR, N_2 adsorption–desorption isotherms, and pH_{pzc} analysis to elucidate its structural, textural, and surface chemical properties. CSAC exhibited a rough and porous morphology, a predominantly amorphous carbon structure, abundant oxygen-containing functional groups, and a micro-mesoporous framework with a specific surface area of 354.54 m^2 g^{-1}. Batch adsorption experiments were conducted to investigate the effects of contact time, pH, and initial TC concentration. Adsorption kinetics were best described by the pseudo-second-order model, indicating chemisorption as the dominant rate-controlling mechanism, while intraparticle diffusion also contributed to the adsorption process. Equilibrium data fitted well to the Langmuir isotherm model, revealing monolayer adsorption with a maximum adsorption capacity of 10.49 mg g^{-1}. The adsorption performance was strongly pH-dependent, with higher removal efficiency under acidic conditions. Regeneration studies demonstrated that CSAC retained good adsorption efficiency over multiple cycles, highlighting its stability and reusability. Overall, this work demonstrates an effective waste-to-resource strategy for producing bio-based activated carbon and offers a sustainable solution for treating antibiotic-contaminated water.

Keywords: *Coconut shell, Activated carbon, Tetracycline, Adsorption, Waste valorization.*

1. INTRODUCTION

The increasing use of antibiotics has become a significant environmental concern because of their continuous release into natural water bodies from pharmaceutical manufacturing effluents, hospital wastewater, livestock farming, and improper disposal of unused medicines [1], [2]. Tetracycline is among the most frequently detected antibiotics in surface water, groundwater, and drinking water sources, due to its extensive use, low biodegradability, and persistence in aquatic environments [3]-[5]. The presence of tetracycline residues poses substantial ecological and public health risks, including the development of antibiotic-resistant bacteria and genes, toxicity to aquatic organisms, and long-term disruption of microbial ecosystems [6], [7]. Conventional wastewater treatment processes are often ineffective at completely removing tetracycline due to its complex chemical structure, multiple ionizable functional groups, and high stability across diverse environmental conditions. Therefore, advanced and effective treatment strategies are urgently needed to address antibiotic contamination. Among the various approaches studied, adsorption has emerged as a promising and practical method because of its simplicity, cost-effectiveness, operational flexibility, and high removal efficiency for a wide range of organic micropollutants, including antibiotics such as tetracycline [8]-[11].

Activated carbon is recognized as an efficient adsorbent for removing antibiotics from water due to its high specific surface area, well-developed porous structure, and abundant surface functional groups. However, its large-scale and sustainable application is constrained by high production costs and dependence on non-renewable precursors such as coal and petroleum-based materials. Recent research has focused on developing activated carbon from biomass waste, which offers a low-cost, renewable, and environmentally friendly alternative [12]-[14]. Agricultural residues, in particular, offer significant advantages due to their abundance, low economic value, and high carbon content. Coconut shells are a promising precursor due to

their high lignin content, inherent hardness, and excellent thermal stability, which facilitate the production of activated carbon with well-developed micro- and mesoporous structures. Converting coconut shell waste into high-value activated carbon reduces environmental burdens associated with biomass disposal and supports waste-to-resource valorization and circular economy principles, thereby contributing to sustainable environmental management.

This study transforms discarded coconut shells into powerful activated carbon, turning agricultural waste into a valuable tool for removing tetracycline from water. By utilizing this sustainable process, the research not only addresses antibiotic pollution but also revitalizes an abundant resource. The activated carbon is thoroughly examined to uncover its unique structure and surface features, while its ability to capture contaminants is rigorously tested. Through detailed analysis of adsorption behavior and mechanisms, the work uncovers the science behind its effectiveness. Ultimately, this research showcases how waste can be reimagined as a solution for cleaner water and a healthier environment.

2. EXPERIMENTS

2.1. Materials and chemicals

Raw coconut shells were collected from local sources in Viet Nam, thoroughly washed with tap water followed by deionized (DI) water to remove adhering impurities, and dried at 105°C for 24 h. Tetracycline hydrochloride (TC, ≥98%) was used as the target antibiotic. Hydrochloric acid (HCl, 37%) and sodium hydroxide (NaOH) were of analytical grade and used without further purification. All solutions were prepared with DI water. The pH of solutions was adjusted using 0.1–1.0 M HCl/NaOH.

2.2. Preparation of coconut shell–derived activated carbon

Dried coconut shells were crushed and sieved to obtain particles sized 0.5–2.0 mm. The material was carbonized in a tubular furnace under a nitrogen atmosphere at a flow rate of 100 sccm. The temperature was increased from room temperature to 650°C at 10°C/min and held for 2 hours. The product was cooled to room temperature, dried at 105°C, and stored in airtight containers. The resulting activated carbon was labeled CSAC.

2.3. Characterization of activated carbon

Surface morphology and elemental composition were analyzed using SEM–EDX. Textural properties, including specific surface area, total pore volume, and pore size distribution, were measured by N_2 adsorption–desorption isotherms at 77 K with the BET method. Surface functional groups were identified by FTIR (4000–400 cm^{-1}). Crystallinity and carbon structure were assessed by XRD (Cu Kα). The point of zero charge (pH$_{pzc}$) was determined using the pH drift method.

2.4. Adsorption of tetracycline by CSAC

Batch adsorption experiments assessed tetracycline (TC) removal using coconut shell–derived activated carbon. A TC stock solution was prepared in deionized water and diluted as needed. Adsorption tests involved mixing a measured amount of adsorbent with the TC solution under controlled agitation and temperature. At regular intervals, samples were filtered, and UV–Vis spectrophotometry at 275 nm was used to measure the remaining TC concentrations. The study systematically examined the effects of pH, contact time, and initial TC concentration. Adsorption capacity and removal efficiency were calculated by mass balance. Kinetic data were analyzed using pseudo-first-order, pseudo-second-order, Bangham, and Elovich models. Equilibrium data were fitted to Langmuir, Freundlich, Elovich, and Temkin isotherms.

2.5. Regeneration and reusability

After adsorption, TC-loaded CSAC was separated and regenerated using ethanol and thermal treatment at 120°C for 120 min, then washed to neutral pH and dried. The regenerated adsorbent was reused for seven cycles under identical conditions to evaluate stability. Regeneration efficiency was reported as the percentage of TC removed in each cycle that was retained after each cycle.

3. RESULTS AND DISCUSSION

3.1. Characterization of CSAC

Figure 1 presents SEM images depicting the surface morphology of coconut shell–derived activated carbon (CSAC) produced by carbonization at 650°C under a nitrogen atmosphere. At low magnification (Figure 1a), CSAC displays a rough, fractured, and heterogeneous surface with numerous cavities and cracks, indicative of biomass decomposition and the release of volatile components during thermal treatment. These characteristics suggest the development of a porous carbon framework

conducive to adsorption. The high-magnification image (Figure 1b) shows that the pores are well-developed and connected, with sizes that are typical of mesoporous structures, ranging from about 450 to 800 nm. This hierarchical porosity facilitates mass transfer and provides accessible adsorption sites, thereby enhancing the uptake of tetracycline molecules from aqueous solutions.
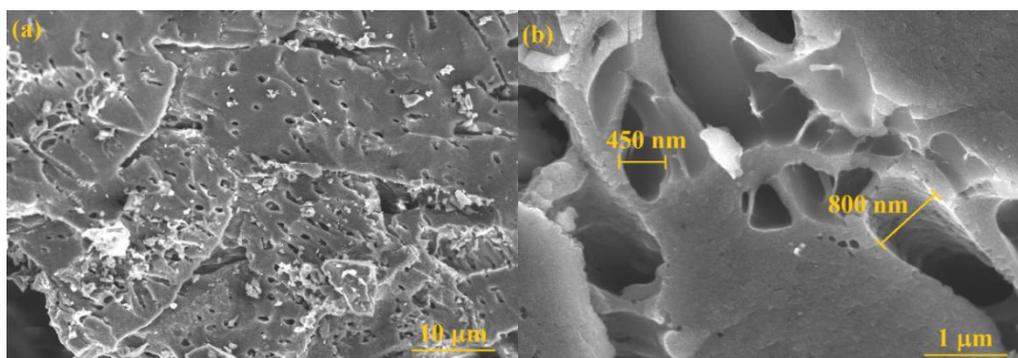
Figure 1. SEM images of CSAC.

The X-ray diffraction (XRD) pattern of CSAC (Figure 2a) shows two broad diffraction peaks at approximately $2\theta \approx 23.3°$ and $43.4°$, corresponding to the (002) and (100) planes of turbostratic carbon, respectively. The lack of sharp crystalline peaks indicates a predominantly amorphous carbon structure, which results from biomass carbonization under an inert atmosphere. This disordered structure enhances adsorption capacity by providing numerous defects and active sites. The Fourier-transform infrared (FTIR) spectrum (Figure 2b) identifies various oxygen-containing functional groups on the CSAC surface, including a broad band near 3400 cm^{-1} attributed to O–H stretching, peaks around 2920–2850 cm^{-1} corresponding to C–H stretching, and bands near 1570–1600 cm^{-1} associated with C=C stretching in aromatic rings. Additional peaks in the 1000–1300 cm^{-1} range indicate C–O stretching vibrations, confirming the presence of surface functionalities that facilitate tetracycline adsorption.
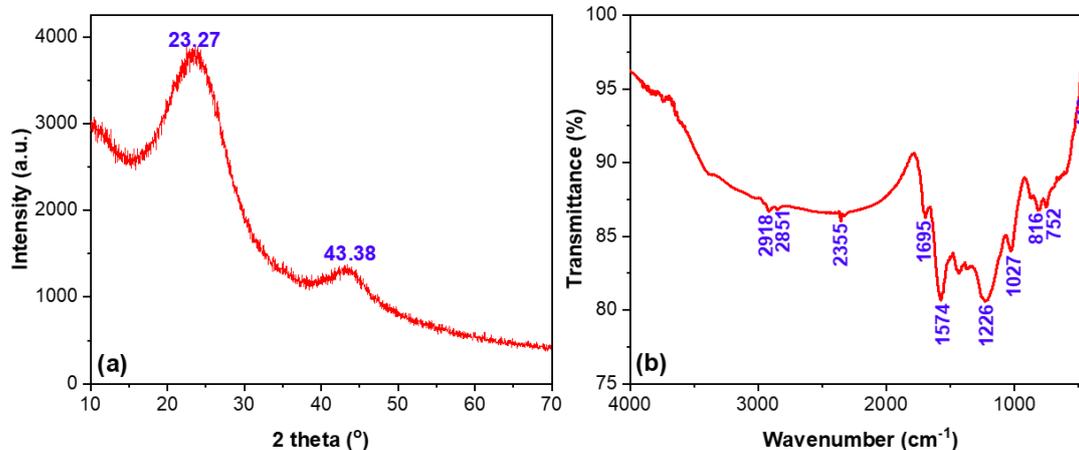
Figure 2. XRD pattern (a) and FTIR spectrum (b) of CSAC.

The EDX spectrum of coconut shell–derived activated carbon (CSAC), presented in Figure 3a, confirms that carbon, comprising 84.39 wt% (88.04 at%), is the predominant element, indicating effective carbonization of the coconut shell precursor. Oxygen is the next most abundant element (14.96 wt%), signifying the presence of oxygen-containing functional groups on the carbon surface that facilitate adsorption processes. Trace amounts of sulfur, chlorine, and calcium are also observed, likely originating from intrinsic biomass constituents or residual inorganic impurities after thermal treatment. The lack of significant metallic impurities further demonstrates CSAC's high purity. These elemental characteristics collectively support the development of a carbon-rich, functionalized surface structure that is well-suited for the efficient adsorption of tetracycline from aqueous solutions.
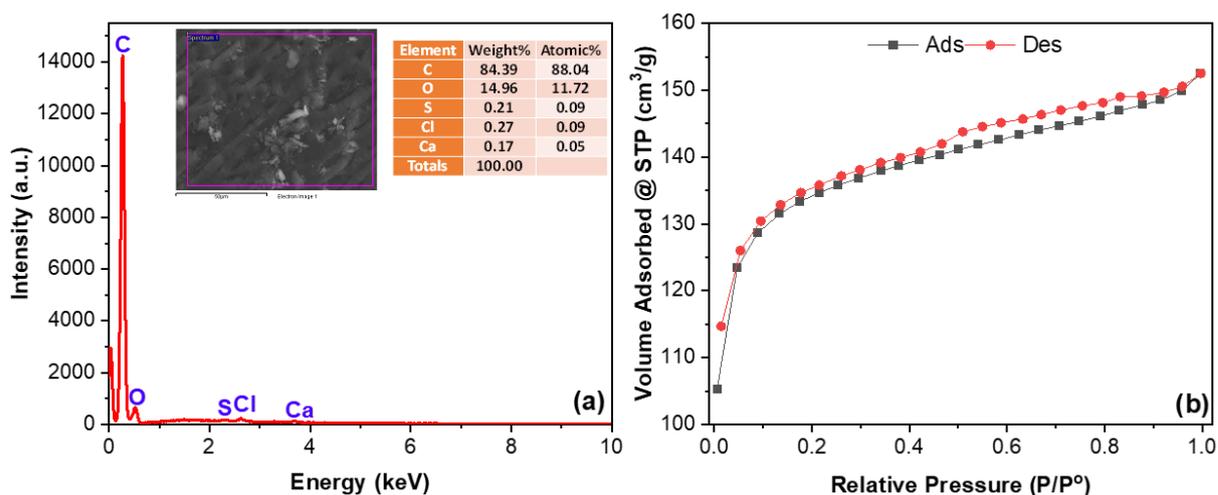
Figure 3. EDX pattern (a) and N_2 adsorption-desorption isotherms of CSAC.

The N_2 adsorption–desorption isotherm of coconut shell–derived activated carbon (CSAC), as presented in Figure 3b, displays a type IV isotherm with a pronounced hysteresis loop, indicating the presence of mesoporous structures. A marked increase in N_2 uptake at low relative pressures ($P/P_0 < 0.1$) indicates micropore contribution, whereas the gradual adsorption at higher P/P_0 values corresponds to mesopore filling. The measured textural parameters, such as a specific surface area of 354.54 m^2 g^{-1}, total pore volume of 0.029 cm^3 g^{-1}, and average pore radius of 1.97 nm, confirm the formation of a micro-mesoporous carbon framework. This hierarchical porosity increases accessible surface area and promotes mass transfer, thereby enhancing the adsorption of organic compounds from aqueous solutions.

3.2. Adsorption of tetracycline by CSAC

The adsorption kinetics of tetracycline (TC) onto coconut shell–derived activated carbon (CSAC) were evaluated using pseudo-first-order (PFO), pseudo-second-order (PSO), Elovich, and Bangham models (Figure 4 and Table 1). Among these, the PSO model shows the best fit, with the highest correlation coefficient ($R^2 = 0.98901$) and a calculated equilibrium adsorption capacity ($q_e = 6.84$ mg g^{-1}) close to the experimental value, indicating that chemisorption is the dominant rate-controlling mechanism. The PFO model exhibits a lower R^2 (0.90691), suggesting limited applicability. The Elovich model ($R^2 = 0.91444$) implies surface heterogeneity and the involvement of energetically diverse active sites. Meanwhile, the excellent fit of the Bangham model ($R^2 = 0.93124$) indicates that pore diffusion also contributes to the adsorption process. Overall, TC adsorption on CSAC proceeds through a combination of surface chemical interactions and intraparticle diffusion.
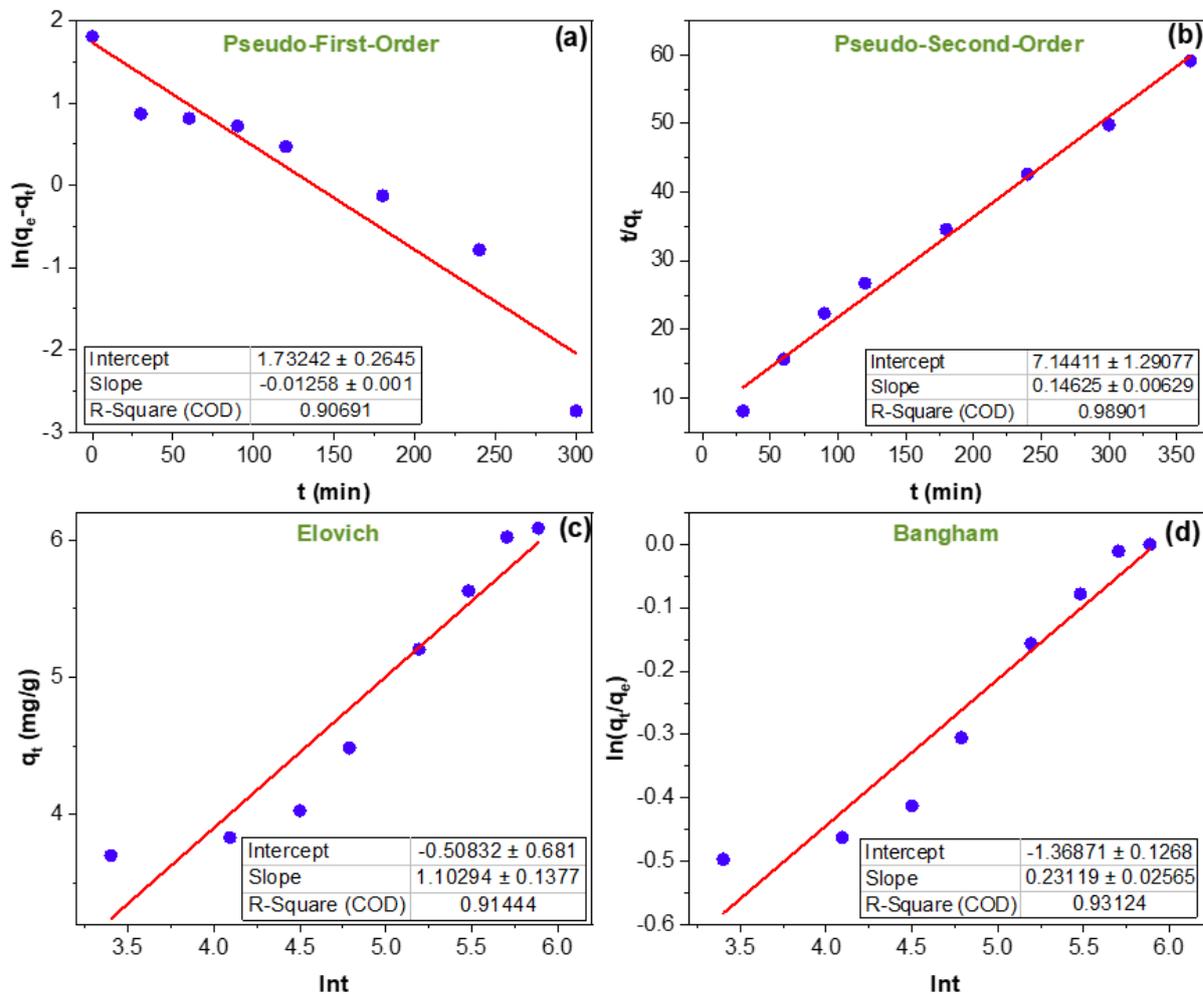
Figure 4. Kinetics adsorption of TC by CSAC: PFO (a), PSO (b), Elovich (c), and Bangham (d).

Table 1. Parameters of kinetic adsorption of TC by CSAC

Models	Parameters	Value	R^2
Pseudo-first order	k_1 (min^{-1})	0.01258	0.90691
	q_e (mg g^{-1})	5.65432	
Pseudo-second order	k_2 (g mg^{-1} min^{-1})	0.00299	**0.98901**
	q_e (mg g^{-1})	6.83761	
Elovich	a (mg g^{-1})	0.69566	0.91444
	b (g mg^{-1})	0.90667	
Bangham	A	0.23119	0.93124
	k_B	0.25444	

The adsorption isotherms of tetracycline (TC) onto coconut shell–derived activated carbon (CSAC) were analyzed using Langmuir, Freundlich, Temkin, and Elovich models (Figure 5 and Table 2). Among them, the Langmuir model provides the best fit with the highest correlation coefficient ($R^2 = 0.99301$), indicating monolayer adsorption on a homogeneous surface with a maximum adsorption capacity (q_{max}) of 10.49 mg g^{-1}. The favorable Langmuir constant ($K_L = 0.3585$ L mg^{-1}) reflects a strong affinity between TC molecules and CSAC. The Freundlich model shows a lower R^2 (0.9208) but an n value of 2.90, suggesting favorable adsorption on a heterogeneous surface. The Temkin model also fits well ($R^2 = 0.9869$), implying that adsorption heat decreases with surface coverage due to adsorbent–adsorbate interactions. The Elovich model presents a reasonable correlation ($R^2 = 0.9528$), further supporting surface heterogeneity. Overall, TC adsorption on CSAC is predominantly monolayer with contributions from heterogeneous interactions.
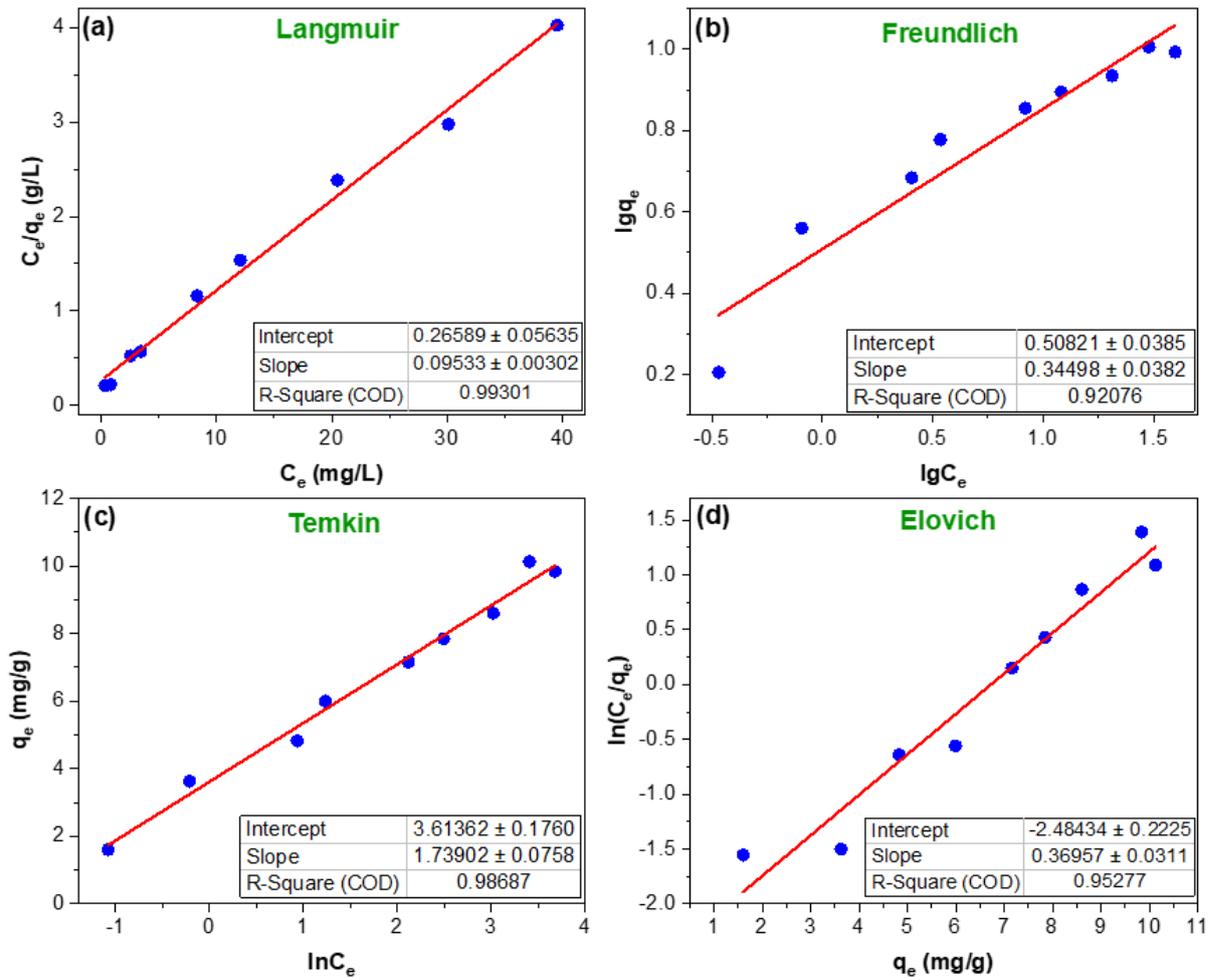
Figure 5. Isotherm adsorption of TC by CSAC: Langmuir (a), Freudlich (b), Temkin (c), and Elovich (d).

Table 2. Parameters of isotherm adsorption of TC by CSAC.

Model	Parameter	Value	R^2
Langmuir	q_{max} (mg g^{-1})	10.4899	0.99301
	K_L (L mg^{-1})	0.3585	
Freundlich	n	2.8987	0.9208
	K_F ((mg g^{-1}).(L mg^{-1})$^{1/n}$)	1.6623	
Temkin	b_T (kJ mol^{-1})	1.4247	0.9869
	K_T	7.9882	
Elovich	q_o (mg g^{-1})	-2.7059	0.9528
	K_E (g mg^{-1})	-0.0308	

Figure 6 illustrates the surface charge behavior of CSAC and the corresponding removal efficiency of tetracycline (TC) as a function of solution pH. As shown in Figure 6a, the point of zero charge (pH$_{pzc}$) of CSAC is approximately 4.62, indicating that the adsorbent surface is positively charged at pH < 4.62 and negatively charged at higher pH values. Figure 6b demonstrates that TC

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removal efficiency decreases markedly with increasing pH, from about 85% under acidic conditions (pH ≈ 3) to around 30% at alkaline pH (≈11). This trend can be attributed to electrostatic interactions between TC species and the CSAC surface, as well as changes in TC speciation with pH. At pH below pH_{pzc}, strong electrostatic attraction enhances adsorption, whereas electrostatic repulsion at higher pH reduces TC uptake. These results highlight the pH-dependent adsorption behavior and confirm the importance of surface charge in governing TC removal by CSAC.
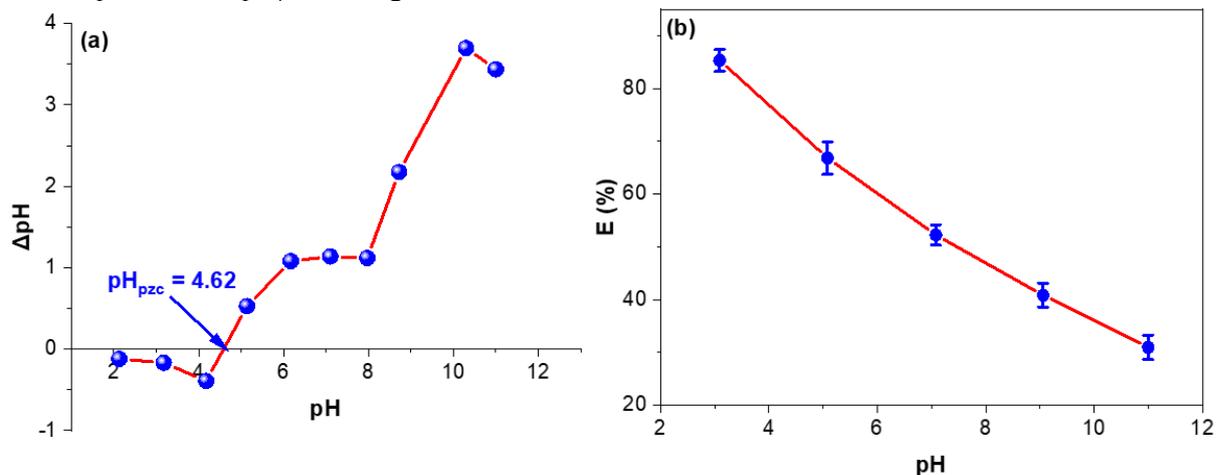

Figure 6. pHpzc (a) and TC removal efficiency (b) with different pH by CSAC.

3.3. Regeneration and reusability

Figure 7 illustrates the time-dependent removal efficiency and reusability of CSAC for tetracycline (TC) adsorption. As shown in Figure 7a, the efficiency of TC removal increases steadily over time, rising from approximately 40% after 30 minutes to approximately 65% after 360 minutes. The rapid initial absorption occurs because many active sites are available on the CSAC surface; however, the rate slows as these sites fill, and it becomes more difficult for TC to diffuse into the CSAC. Figure 7b demonstrates the recyclability of CSAC over five consecutive adsorption cycles. Although a slight decrease in removal efficiency is observed, from around 67% in the first cycle to about 62% after the fifth cycle, CSAC maintains relatively stable performance. This result shows that CSAC is highly stable and can be reused effectively, highlighting its usefulness for cleaning water contaminated with antibiotics.
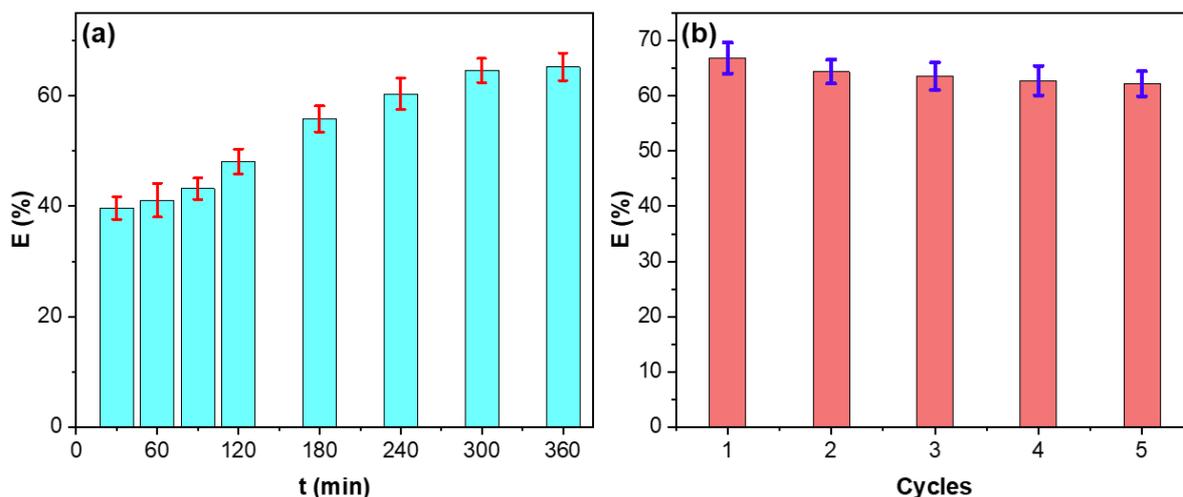

Figure 7. Removal efficiency of TC vs. time (a) and after 5 cycles (b) on CSAC.

4. CONCLUSION

This study successfully demonstrates the waste-to-resource conversion of coconut shells into activated carbon for the effective removal of tetracycline from aqueous solutions. The prepared CSAC exhibited a porous and heterogeneous carbon structure with abundant surface functional groups and favorable textural properties, which collectively contributed to its

adsorption performance. Adsorption kinetics followed a pseudo-second-order model, indicating that chemisorption dominated the removal process, with pore diffusion also playing a significant role. The Langmuir isotherm provided the best description of equilibrium behavior, confirming monolayer adsorption with a maximum capacity of 10.49 mg g^{-1}. The adsorption efficiency was strongly influenced by solution pH, with enhanced TC uptake under acidic conditions due to favorable electrostatic interactions. Importantly, CSAC maintained a relatively stable removal efficiency across repeated regeneration cycles, indicating good structural stability and reusability. These findings emphasize the attractiveness of coconut shell–derived activated carbon as a low-cost, sustainable, and environmentally friendly adsorbent, offering a promising approach for mitigating antibiotic pollution while promoting biomass waste valorization and circular economy principles.

DECLARATION OF CONFLICTS OF INTEREST

The authors declare that this research has no conflicts of interest.

ACKNOWLEDGMENTS

REFERENCES

[1] V. O. Ajekiigbe *et al.*, "The increasing burden of global environmental threats: role of antibiotic pollution from pharmaceutical wastes in the rise of antibiotic resistance," *Discover Public Health,* vol. 22, no. 1, pp. 1-10, 2025.

[2] S. Zampelli and R. Verna, "Environmental Pollution from Pharmaceuticals," *Life,* vol. 15, no. 9, p. 1341, 2025.

[3] L.-J. Zhou *et al.*, "Trends in the occurrence of human and veterinary antibiotics in the sediments of the Yellow River, Hai River and Liao River in northern China," *Environmental Pollution,* vol. 159, no. 7, pp. 1877-1885, 2011.

[4] Z. Wang *et al.*, "Characterization and source identification of tetracycline antibiotics in the drinking water sources of the lower Yangtze River," *Journal of environmental management,* vol. 244, pp. 13-22, 2019.

[5] X. Chen, Y. Yang, Y. Ke, C. Chen, and S. Xie, "A comprehensive review on biodegradation of tetracyclines: Current research progress and prospect," *Science of The Total Environment,* vol. 814, p. 152852, 2022.

[6] Y. Amangelsin, Y. Semenova, M. Dadar, M. Aljofan, and G. Bjørklund, "The impact of tetracycline pollution on the aquatic environment and removal strategies," *Antibiotics,* vol. 12, no. 3, p. 440, 2023.

[7] Y. Wang, B. Du, and G. Wu, "Tetracycline in anaerobic digestion: Microbial inhibition, removal pathways, and conductive material mitigation," *Journal of Hazardous Materials,* vol. 496, p. 139378, 2025.

[8] L. Perelomov *et al.*, "Adsorption of antibiotics by natural clay minerals," *Minerals,* vol. 15, no. 7, p. 733, 2025.

[9] Z. Hajyani, Z. Mousavi, M. Soleimanbeigi, Y. J. Wong, and A. A. Beni, "Antibiotic adsorption from real pharmaceutical factory wastewater using a hole-punched flat ceramic membrane based on clay and hydroxyapatite in a fixed bed module," *Results in Engineering,* vol. 26, p. 105364, 2025.

[10] J. Zhang *et al.*, "Adsorption of tetracycline by polycationic straw: Density functional theory calculation for mechanism and machine learning prediction for tetracyclines' remediation," *Environmental Pollution,* vol. 340, p. 122869, 2024.

[11] P. Hongsawat, P. Kalnaowakul, and P. Prarat, "Water-based Synthesis of MIL-53 (Al) at Room Temperature: An Effective Adsorbent for Oxytetracycline (OTC) Antibiotic Removal from Water," *CHIANG MAI JOURNAL OF SCIENCE,* vol. 51, no. 1, 2024.

[12] M. Gayathiri, T. Pulingam, K. Lee, and K. Sudesh, "Activated carbon from biomass waste precursors: Factors affecting production and adsorption mechanism," *Chemosphere,* vol. 294, p. 133764, 2022.

[13] Suhas *et al.*, "Transforming Biomass Waste into Hydrochars and Porous Activated Carbon: A Characterization Study," *Resources,* vol. 14, no. 3, p. 34, 2025.

[14] S. Ullah *et al.*, "Activated carbon derived from biomass for wastewater treatment: Synthesis, application and future challenges," *Journal of Analytical and Applied Pyrolysis,* vol. 179, p. 106480, 2024.

Nghiên cứu chuyển hóa nguồn thải gáo dừa thành than hoạt tính ứng dụng loại bỏ tetracycline

TÓM TẮT

Sự gia tăng dư lượng kháng sinh trong môi trường nước đã gây ra những lo ngại nghiêm trọng về sinh thái và sức khỏe cộng đồng. Trong nghiên cứu này, vỏ dừa đã được chuyển hóa thành than hoạt tính (CSAC) thông qua quá trình cacbon hóa nhiệt và được ứng dụng như một chất hấp phụ giá rẻ và bền vững để loại bỏ tetracycline (TC) khỏi dung dịch nước. Các kết quả SEM–EDX, XRD, FTIR, BET và pHpzc đã được sử dụng để đặc trưng cấu trúc và tính chất bề mặt của CSAC điều chế được. CSAC có dạng xốp, cấu trúc carbon chủ yếu là vô định hình, có nhiều nhóm chức chứa oxy và khung trung-vi xốp với diện tích bề mặt riêng là 354,54 m^2 g^{-1}. Các thí nghiệm hấp phụ theo mẻ đã được tiến hành để nghiên cứu ảnh hưởng của thời gian tiếp xúc, pH và nồng độ TC ban đầu. Quá trình hấp phụ được mô tả tốt nhất bằng mô hình động học giả bậc hai và phù hợp tốt với mô hình đẳng nhiệt Langmuir (dung lượng hấp phụ tối đa là 10,49 mg g^{-1}). Hiệu suất hấp phụ phụ thuộc mạnh vào pH, hiệu quả loại bỏ TC đạt cao hơn trong môi trường axit. Các nghiên cứu tái sinh chất hấp phụ cho thấy CSAC duy trì hiệu quả hấp phụ tốt qua 5 chu kỳ, cho thấy sự ổn định và khả năng tái sử dụng của CSAC. Nghiên cứu này chứng minh một chiến lược chuyển đổi chất thải thành nguyên liệu hiệu quả để sản xuất than hoạt tính sinh học và cung cấp một giải pháp bền vững để xử lý nước bị ô nhiễm kháng sinh.

Từ khoá: Gáo dừa, Than hoạt tính, Tetracycline, Hấp phụ, Tận thu chất thải.