



Development of High-performance Activated Carbon Derived from Longan Peel for Caffeine Removal in Ion-rich Hospital Wastewater

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Abstract: This study presents the development of activated carbon derived from longan peel (LPAC) as an efficient and low-cost adsorbent for the removal of caffeine (CAF) from synthetic hospital wastewater. Longan peel was carbonized and chemically activated at 600 °C and 700 °C, producing LPAC-600 and LPAC-700, respectively. The structural, textural, and functional properties of the adsorbents were characterized using XRD, FTIR, N₂ adsorption-desorption analysis, and SEM. The results indicated that increasing the activation temperature significantly enhanced surface area, pore volume, and graphitic carbon structure, with LPAC-700 exhibiting the highest BET surface area (1401.8 m²/g) and mesoporosity. Batch adsorption experiments demonstrated that LPAC-700 achieved superior CAF removal compared to LPAC-600 in both deionized water and synthetic hospital wastewater, although ion-rich conditions reduced removal efficiency due to competitive adsorption. The optimal adsorbent dosage was 0.3 g/L, yielding up to 94.23% CAF removal at an initial concentration of 100 ppm. Solution pH strongly influenced adsorption, with maximum removal at pH 7, attributed to favorable π - π interactions and hydrogen bonding. These findings confirm that LPAC-700 is a promising bio-derived adsorbent for caffeine removal in complex wastewater systems.

Keywords: Activated carbon; longan peel; caffeine; synthetic hospital wastewater

1. Introduction

Caffeine (CAF) is among the most widely ingested bioactive compounds worldwide, functioning as a central nervous system stimulant and appearing in a broad spectrum of commercial products, including beverages, functional foods, dietary supplements, and pharmaceutical formulations [1-3]. Because of its extensive global consumption and relatively low metabolic transformation in humans, a significant proportion of ingested caffeine is excreted unmetabolized. Consequently, CAF is increasingly observed in natural and engineered aquatic environments, where it is now recognized as an emerging anthropogenic contaminant of environmental relevance. Regulatory agencies, including the United States Environmental Protection Agency (EPA), have reported that caffeine serves as a reliable marker for domestic and sanitary wastewater contamination, providing insight into the performance and integrity of water treatment systems [4].

Concerns over caffeine's ecological implications, such as its potential effects on aquatic organisms, persistence in water matrices, and co-occurrence with other pharmaceutical residues, have driven ongoing efforts to develop efficient removal technologies. Wastewater originating from hospitals frequently contains relatively high concentrations of caffeine, partly due to the extensive use of CAF-containing medications and diagnostic agents [4-6]. Moreover, such effluents generally exhibit elevated ionic strength, which adds additional complexity to the removal processes. High ionic concentrations can interfere with adsorption or catalytic reactions by competing for active sites or altering surface charge interactions, making remediation of caffeine in these matrices particularly challenging. Previous investigations have reported that CAF removal can be achieved using advanced oxidation systems, ozonation, membrane-based separations, and adsorption-based technologies; however, limitations in operational cost, energy consumption, and maintenance often restrict their practical application [7-9].

Among the various technologies, adsorption remains an especially promising approach for removing caffeine and other pharmaceutical contaminants. This method offers multiple advantages, including low operational cost, ease of implementation, compatibility with existing treatment infrastructures, and minimal generation of harmful by-products. A variety of adsorbent materials, such as zeolites, activated carbon, mesoporous silica materials, and metal-organic frameworks (MOFs), have been examined for their capacity to remove pharmaceuticals from water [9-12]. Despite their effectiveness, many of these materials have drawbacks, including high production costs, difficulty in regeneration, or limited sustainability. Activated carbon, in contrast, is a well-established adsorbent widely used across industrial sectors due to its high porosity, tunable surface chemistry, and substantial adsorption capacity for organic and inorganic pollutants [6,10]. Nonetheless, reliance on conventional raw materials and the challenges associated with separating powdered activated carbon from aqueous systems have prompted growing interest in alternative biomass-derived sources. Agricultural waste provides a renewable, abundant feedstock for producing cost-efficient activated carbon while contributing to waste valorization. Longan peel, rich in carbonaceous content and widely available in Southeast Asia, has recently emerged as a promising precursor for generating activated carbon with desirable structural properties [13].

The present study investigates the feasibility of producing activated carbon derived from longan peel (LPAC) through chemical activation at different temperatures, to assess its performance in removing caffeine from synthetic hospital wastewater. Comprehensive structural, functional, and morphological characterization techniques, including XRD, FTIR, N₂ adsorption-desorption analysis, and SEM, were employed to elucidate the influence of activation temperature on the physicochemical properties of the prepared materials. Additionally, the stability of LPAC after exposure to caffeine-containing solutions was examined to determine its potential as a robust, sustainable bioadsorbent. By evaluating LPAC performance in a highly ionized wastewater system, this work aims to establish the viability of longan peel-derived activated carbon as an environmentally friendly, economically feasible candidate for advanced wastewater treatment applications.

2. Materials and Methods

2.1 Chemicals and reagents for preparation of longan peel activated carbon (LPAC)

Sodium hydroxide (NaOH; analytical grade 99%) was acquired from Sigma-Aldrich. All chemical solutions were prepared using deionized (DI) water from RCI Labscan, which was also used to rinse and clean the samples. Longan peel, a common agricultural residue material, was collected from Rim Rong Housewives Community Enterprise, Makhuea Chae Subdistrict, Mueang District, Lamphun Province, Thailand.

2.2 Preparation of longan peel activated carbon (LPAC)

The following process was used to produce the longan peel activated carbon. Before being stored in silica gel desiccators, the raw longan peel was crushed and sieved to achieve a particle size of 100 mesh. Then, it was dried in an oven at 100°C for 10 h to remove any water absorbed on the surface. The dried longan peel was carbonized for 1.5 h at 400 °C. After cooling to room temperature, the resulting carbon was crushed. Sodium hydroxide (NaOH) was combined with the carbonized material in a 1:2 (w/w) ratio. The mixture was

then activated at 600 and 700 °C for 1.5 h. The resulting activated carbons from the longan peel were labeled as LPAC-600 and LPAC-700, respectively. Then, they were cooled to room temperature and washed with distilled water until their pH reached 7. The LPAC materials were then dried in a hot air oven at 80 °C for 24 h. The yield of LPAC-600 and LPAC-700 was estimated to be in the range of 18–25 wt%, which is consistent with previously reported values for NaOH-activated longan seed-derived carbons prepared at high temperatures [14].

2.3 Characterization of adsorbents

Structural information about the materials was obtained by powder X-ray diffraction (XRD) using an Empyrean 3 Panalytical diffractometer with Cu K α radiation at 40 kV and 40 mA. N₂ adsorption-desorption measurements were performed at -196 °C utilizing an Anton Paar Nova 600 to determine the textural properties of the materials. The sample weight, initially approximately 40 mg, was precisely measured following pretreatment at 150 °C for 2 hours. Fourier-transform infrared spectroscopy (FTIR) was utilized to identify functional groups in the composites, employing a Perkin-Elmer spectrometer with the KBr technique. Sixty-four scans were averaged for each spectrum, covering the range of 400–4000 cm⁻¹ at a resolution of 4 cm⁻¹. The morphology of the sample was assessed using scanning electron microscopy (SEM). The SEM images were captured using a TESCAN VEGA3 scanning electron microscope functioning at 10 kV. The samples on carbon tape were examined with a gold coating applied.

2.4 Preparation of synthetic hospital wastewater (HP wastewater)

The synthetic hospital effluent was prepared to represent a substantial volume of ionized wastewater, following the methodology reported by Tiwari et al. (2019) [15]. The mixture comprised (NH₄)₂SO₄ at 353.57 ppm, KH₂PO₄ at 92.19 ppm, MgSO₄ at 34.68 ppm, CaCl₂·2H₂O at 22.10 ppm, FeCl₃ at 11.63 ppm, CuSO₄ at 0.07 ppm, Na₂MoO₄·2H₂O at 0.10 ppm, MnSO₄·H₂O at 0.12 ppm, ZnCl₂ at 0.23 ppm, CoSO₄·7H₂O at 0.48 ppm, Na₂CO₃ at 428.60 ppm, and C₆H₁₂O₆ at 938.35 ppm. Furthermore, 100 ppm of caffeine (C₈H₁₀N₄O₂, CAF) was incorporated into the synthetic hospital wastewater. The chemicals utilized were of analytical grade and obtained from Sigma-Aldrich. A batch adsorption study was performed for 48 hours at 150 rpm and room temperature. The samples passed filtration, and the total removal was determined by analyzing the UV-Vis spectra over 200 to 800 nm.

2.5 Adsorption experiment

Adsorption studies of CAF on longan peel-activated carbon were conducted in a batch system using 50-mL Erlenmeyer flasks containing CAF solution and dried adsorbents. The adsorption experiments were performed by shaking the flasks in a shaker under specified conditions. The concentration of the CAF solution post-adsorption was quantified using a UV-vis spectrophotometer at 273.5 nm. The characteristic absorption of caffeine, and no significant interference was observed. Additionally, calibration using standard solutions ensures reliable quantification. The amount of CAF removal and adsorption capacity was calculated according to a mass balance on the CAF concentration using Eq (1) and Eq. (2), respectively:

$$\% \text{ CAF Removal} = \frac{(C_i - C_e)}{C_i} \times 100 \quad (1)$$

$$Q_e = \frac{(C_i - C_e)}{W} \times V \quad (2)$$

where C_i is the initial concentration of CAF (ppm, mg/L), C_e is the equilibrium concentration of CAF in solution (ppm, mg/L), Q_e is the adsorption capacity at adsorption equilibrium (mg/g, dried adsorbent), V is the volume of initial solution (L), and W is the weight of dried adsorbent (g). All experiments were conducted in duplicate, and the reproducibility of the data was verified with deviations within an acceptable range (<5%).

3. Results and Discussion

3.1 Characterization of LPAC adsorbents

Figure 1 presents the XRD patterns of activated carbon derived from longan peel, specifically LP-600 and LP-700. Two distinct peaks were observed in both materials. The prominent peak observed at 15–30° is

likely attributable to reflection plane C(002), indicative of a disordered aromatic carbon structure, while the peak at 43.96° corresponds to reflection plane C(100/101) associated with graphitic or organized carbon [16]. The signal intensity of the graphitic carbon in LPAC-700 was significantly greater than that in LPAC-600, suggesting that the elevated activation temperature led to an improvement in the graphitic carbon structure.

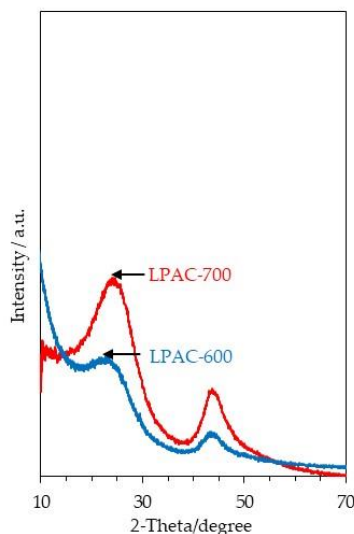


Figure 1. XRD patterns of LPAC adsorbents.

The FT-IR analysis provided information for identifying the chemical functional groups in longan peel activated carbon at different activation temperatures. Figure 2 shows the FTIR spectra of LPAC-600 and LPAC-700 materials. The broad band at approximately 3440 cm^{-1} was attributed to the O-H stretching of hydroxyl groups and adsorbed water. The C-H stretching vibration characteristic of the alkyl structure in the aliphatic group was observed at 2920 cm^{-1} [17]. The band at 1630 cm^{-1} could be assigned to C-C vibrations in aromatic rings and C=O stretching vibrations of carboxyl and carbonyl groups. The bands within the range of $1000 - 1300\text{ cm}^{-1}$ were attributed to oxidized carbons, specifically the C-O stretching vibrations associated with carboxylic acids, alcohols, phenols, ethers, and esters [17,18]. The intensity of the LPAC-700 peak was observed to be lower than that of the LPAC-600 peak. The results indicated that higher activation temperatures led to the decomposition of these organic compounds [19].

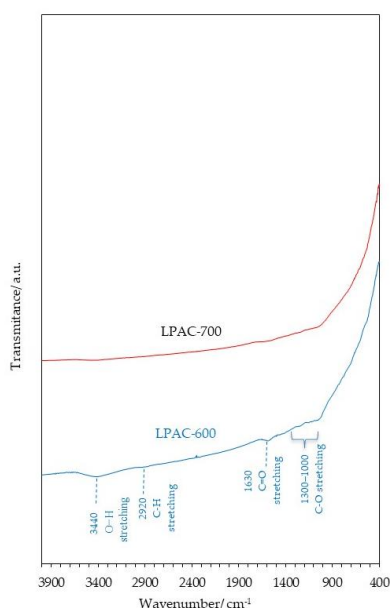


Figure 2. FT-IR spectra of LPAC adsorbents.

Figure 3 shows the N₂ adsorption–desorption isotherms and pore size distributions of LPAC materials. According to IUPAC, the isotherms of these materials could be classified as type IV, which are characteristic of materials with well-developed mesoporous structures. The LPAC-700 exhibited a higher N₂ adsorbed volume than the LPAC-600 (Figure 3A), corresponding to the specific surface area of these materials, as shown in Table 1. These results indicated that the activation temperature resulted in the textural properties of the activated carbon. In addition, the pore size distribution of LPAC materials, as shown in Figure 3b, indicated that most pores were distributed in the 2–10 nm size range, confirming the material's mesoporosity. However, the LPAC-600 exhibited a lower pore volume and average pore diameter (Table 1) than that of the LPAC-700. These results suggested that increasing the activation temperature resulted in distinct pore structures.

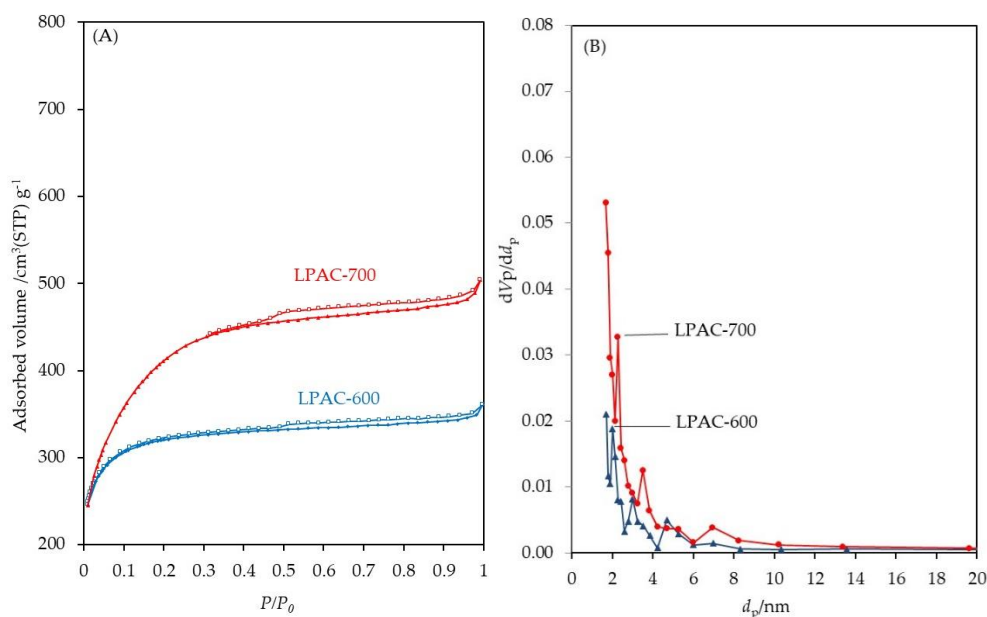


Figure 3. N₂ adsorption-desorption isotherms (A) and BJH pore size distribution (B) of LPAC adsorbents.

Table 1. Textural properties of LPAC adsorbents.

Sample	Specific surface area (m ² /g)	Pore volume (cm ³ g ⁻¹)	Average Pore diameter (nm)
LPAC-600	1004.5	0.66	8.5
LPAC-700	1401.8	0.82	9.2

3.2 Caffeine adsorption studies

In the present study, we investigated how different types of activated carbons affect the CAF adsorption process using an initial concentration of 100 ppm of CAF dissolved in DI water and in synthetic hospital wastewater (representative of high ionized wastewater), as shown in Figure 4. The LPAC adsorbents were examined for their effectiveness in removing CAF from aqueous solutions, both with and without the ion present, as illustrated in Figures 4A and 4B, respectively. The adsorption conditions included an adsorbent/solution (M/V) ratio of 0.1 g/L, a CAF concentration of 100 ppm, a time duration of 120 minutes, a shaking speed of 120 rpm, and room temperature. The longan peel activated carbon produced at higher activation temperatures showed a higher percentage of CAF removal (LPAC-600 < LPAC-700), which correlated with BET surface area and pore volume, as shown in Table 1. The findings indicated that the highest CAF removal was contingent on the adsorbent's textural properties. Consequently, the LPAC-700 proved to be an appropriate adsorbent for subsequent investigations. However, the CAF removal from synthetic hospital wastewater with high ion concentrations showed lower adsorption for both adsorbents, as shown in Figure 4B.

A significant decrease in CAF removal was observed in synthetic hospital effluent compared with deionized water, indicating the influence of coexisting ions on adsorption efficacy. This phenomenon cannot be ascribed solely to mere competition but also encompasses other physical factors. Multivalent cations,

including Ca^{2+} and Mg^{2+} , may compete with CAF molecules for adsorption sites, especially those linked to oxygen-containing functional groups. Moreover, increased ionic strength results in electrostatic screening, diminishing the effective interaction between caffeine molecules and the adsorbent surface. Furthermore, hydration shells encasing dissolved ions may impede the diffusion of CAF into porous matrices, consequently diminishing adsorption efficiency [6]. These results were confirmed by comparing FTIR spectra obtained before and after caffeine adsorption in synthetic wastewater, as shown in Figure 5. The FTIR spectra of LPAC after adsorption showed multiple peaks corresponding to caffeine molecules adsorbed on its surface.

In addition, the synthesized activated carbons exhibited an adsorption capacity for CAF comparable to that of the commercial activated carbon (AC) with a BET surface area of $1200 \text{ m}^2/\text{g}$, reported by Rajkamal et al. to be around 71.7 mg/g , depending on operational conditions and surface properties [20]. Furthermore, Figure 6 shows SEM images of LPAC materials before and after CAF adsorption in synthetic HP wastewater. The LPAC exhibited a particle size of $50\text{--}125 \text{ nm}$ and a high porosity. After adsorption of CAF in synthetic HP wastewater, the LPAC showed no difference in morphology from the initial.

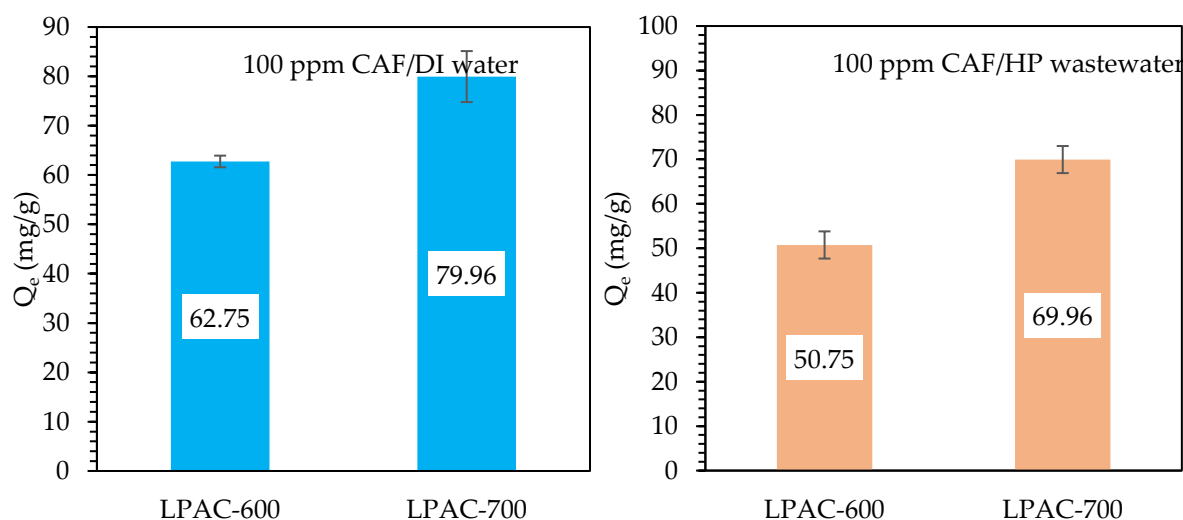


Figure 4. Effect of type of LPAC adsorbents on CAF removal in (A) DI water, (B) synthetic HP wastewater.

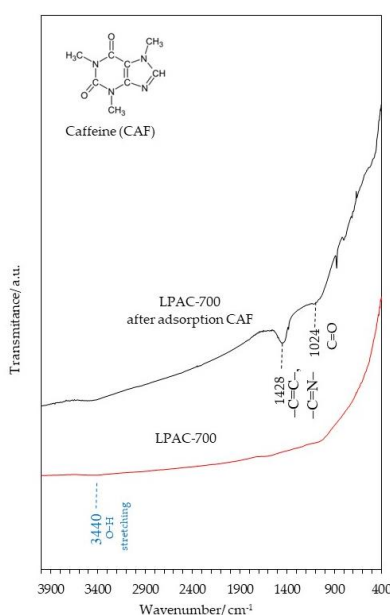


Figure 5. FT-IR spectra of LPAC-700 before and after adsorption of CAF in synthetic HP wastewater.

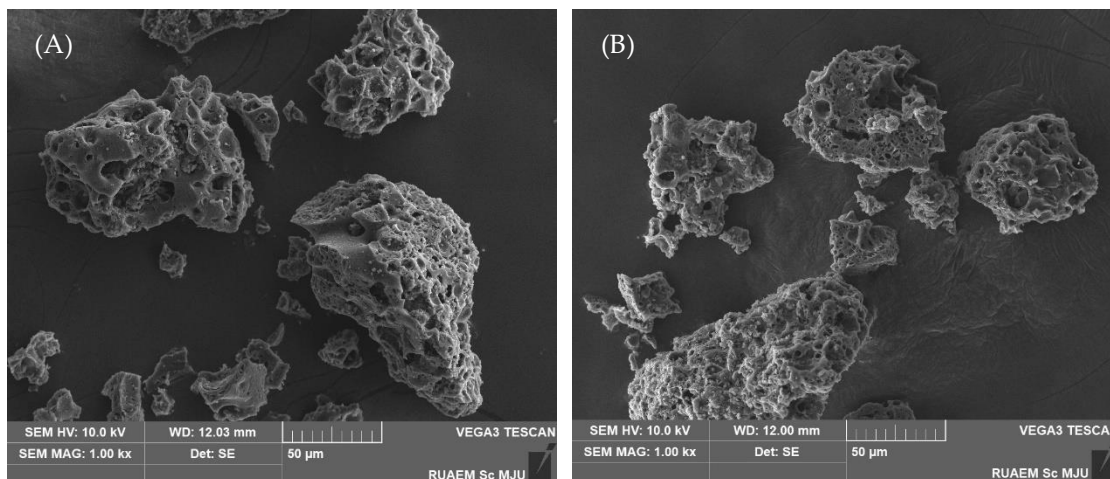


Figure 6. SEM images of LPAC materials (A) before and (B) after adsorption of CAF in synthetic HP wastewater.

Figure 7 illustrates the impact of the mass of adsorbent per volume of CAF solution (M/V) on the removal of CAF by LPAC-700. The adsorption conditions comprised an adsorbent/solution (M/V) ratio of 0.1–0.3 g/L, a CAF concentration of 100 ppm, a duration of 120 minutes, a shaking speed of 120 rpm, and ambient temperature. Increasing M/V from 0.1 g/L to 0.3 g/L resulted in an increase in CAF removal percentage from 69.96 to 94.23. An increase in the mass of adsorbent per volume led to a corresponding increase in pore volume and the presence of functional groups ($-\text{COOH}$, $-\text{OH}$, and aliphatic $-\text{C}=\text{C}-$) in the material. The physicochemical adsorption capacity of this adsorbent for CAF removal was superior. The results indicate that 0.3 g/L is the optimal mass-to-volume ratio for treating 1 L of CAF solution at 100 ppm.

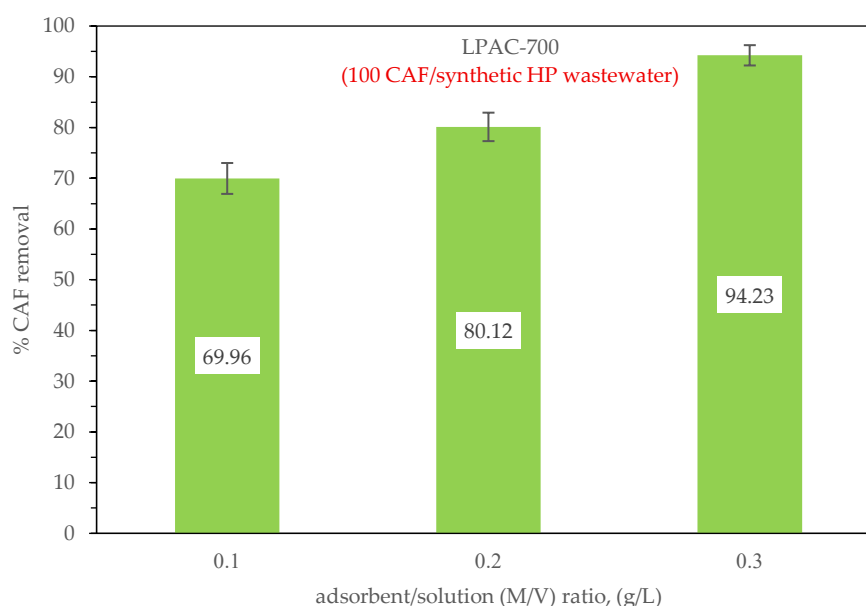


Figure 7. Effect of LPAC adsorbent dose on CAF removal.

The pH of a solution significantly affects adsorption processes by influencing the interactions between adsorbents and adsorbates. Variations in pH can alter the surface charge of adsorbents and affect the protonation state of functional groups on the adsorbent surface. The effect of pH on the CAF removal of LPAC-700 are shown in Figure 8. The adsorption conditions were as follows: pH range of 4–10, adsorbent/solution (M/V) ratio of 0.3 g/L, CAF concentration of 100 ppm, duration of 120 minutes, shaking speed of 120 rpm, and maintained at room temperature. The findings indicate that the maximum percentage of CAF removal was

achieved at pH 7, with a notable reduction at pH 10. The findings align with those of Beltrame et al. [10], who observed a significant pH change during the removal of CAF utilizing mesoporous activated carbon fibers derived from pineapple plant leaves. At pH 7, the predominance of the neutral form of CAF and the activated carbon's negative surface charge created optimal conditions for CAF removal. Many processes influence the adsorption of CAF onto LPAC. The aromatic configuration of CAF facilitates π - π interactions with the graphitic domains of LPAC, which are more evident in LPAC-700 owing to increased graphitization. Furthermore, mesopores (2–10 nm) correspond well with the molecular dimensions of CAF (~0.8 nm), promoting pore-filling adsorption [18,19]. The increase in solution pH led to a decrease in CAF removal, indicating repulsion between CAF molecules and the negatively charged surface of the activated carbon [10].

Although full isotherm modeling was not conducted, the adsorption behavior is expected to follow Langmuir-type monolayer adsorption due to the activated carbon's homogeneous surface, as reported in similar systems. Furthermore, CAF adsorption on activated carbon is commonly described by pseudo-second-order kinetics, consistent with the chemisorption mechanisms reported by Rajkamal et al. [20]. However, these materials will be further studied for their adsorption behavior and kinetics compared to commercially available activated carbon. Importantly, this study demonstrates that LPAC maintains reasonable adsorption efficiency even under ion-rich conditions, which are representative of real hospital wastewater. This highlights its potential applicability in practical wastewater treatment systems.

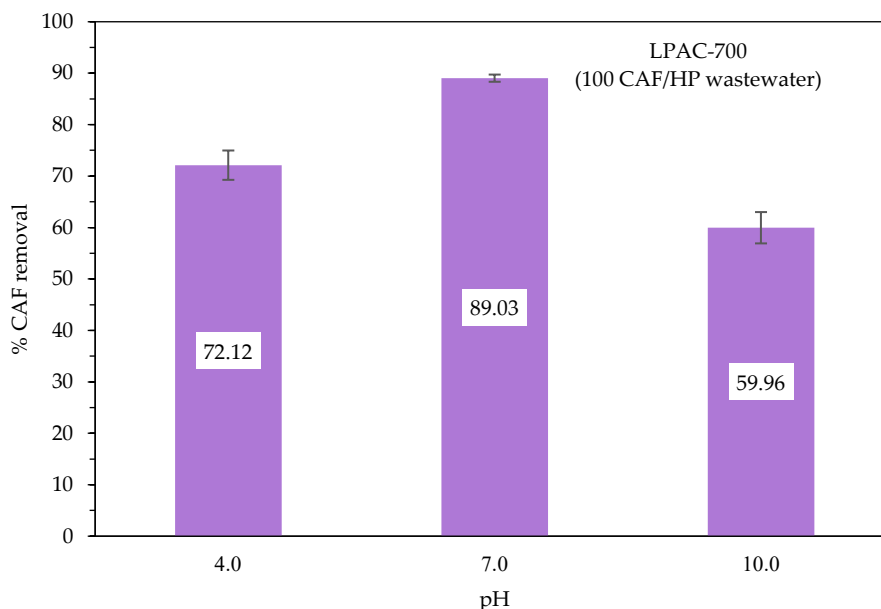


Figure 8. Effect of pH on CAF removal.

4. Conclusions

Activated carbon produced from longan peel demonstrated high potential for removing caffeine from aqueous systems, particularly LPAC-700, which offered superior textural and structural properties due to higher activation temperature. The mesoporous structure, large specific surface area, and suitable functional groups contributed to its high adsorption efficiency. Although ion-rich synthetic hospital wastewater reduced caffeine adsorption through competitive interactions, LPAC-700 still maintained substantial removal capability. Adsorbent dosage and solution pH were critical parameters, with optimal performance observed at 0.3 g/L and neutral pH. Overall, longan peel-derived activated carbon is an effective, sustainable, and inexpensive adsorbent, demonstrating its applicability for treating pharmaceutical-contaminated wastewater in real-world settings.

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Conflicts of Interest: The authors declare no conflict of interest.

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