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Sustainable solutions for pharmaceutical contamination: Potassium hydroxide and phosphoric acid modified biosorbents from agricultural waste and coconut shells for acetaminophen removal

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ABSTRACT

This research explores the utilization for adsorbents produced from agricultural waste (AW) and coconut shells (CS), that were chemically activated through the use of phosphoric acid (H_3PO_4) and potassium hydroxide (KOH), to evaluate their efficacy in eliminating acetaminophen (ACT) from water. Batch tests were carried out to assess the impact of operational parameters, including pH and adsorbent dose, initial ACT concentration, and contact time, on adsorption performance. Under optimal conditions – including a dose of 1.1 g/L, pH 8.0 for all modified adsorbents (except AW-H_3PO_4 at pH 4), a contact duration of 120–150 min, and ACT concentrations of 100 and 120 mg/L – a highest effectiveness of removing of 95% was attained.. Chemical modification of the raw materials considerably enhanced the surface area of the adsorbents, improving their adsorption capabilities. Adsorption behavior was further examined utilizing a variety of isotherm and kinetic models, with the pseudo-second order model providing the most accurate fit. Isotherm results indicated monolayer adsorption on heterogeneous surfaces. Langmuir analysis revealed a maximum capacity for adsorption of 10.07 mg/g of AW-H_3PO_4 and 9.699 mg/g for AW-KOH, surpassing the 5.102 mg/g capacity of unmodified agricultural waste. Similarly, modified coconut shell adsorbents showed improved capacities – 11.416 mg/g for CS-H_3PO_4 and 10.549 mg/g for CS-KOH, compared to 4.831 mg/g for the unmodified one.

Keywords: agricultural waste; coconut shell waste; adsorption; acetaminophen; low-cost adsorbent; chemical activation

INTRODUCTION

Water stands as one of the most vital natural resources on the planet, yet its quality continues to decline over time, largely due to increasing human impact, rapid population growth, industrial development, and unchecked exploitation of natural assets (Jabeen et al., 2015). As a result, safeguarding and enhancing water quality have become increasingly urgent. Among the most pressing environmental issues today is water pollution, particularly from emerging contaminants such as pharmaceutical residues, which present a significant global threat (Irshad et al., 2023; Lu et al., 2025). This growing concern calls for sustainable and effective strategies to eliminate these pollutants and ensure the protection of water resources (Letsoalo et al., 2023). Pharmaceutical substances – including common anti-inflammatory drugs like diclofenac, ibuprofen, and ketoprofen – have garnered attention due to their persistence in the environment (Silva et al., 2022). Commonly employed in both Veterinary and human medicine to treat or prevent infections, these compounds are frequently detected in a range of aquatic systems, from surface and groundwater to wastewater and even handled water for drinking (Mohammadi et al., 2022; Yan et al., 2022). Their presence can

be traced back to various sources, including effluents from pharmaceutical manufacturing, hospitals, and biological waste from both humans and animals (Gu et al., 2025; Khmaissa et al., 2024). What makes these substances particularly problematic is their key characteristics: limited biodegradability, significant toxicity, and potential to induce mutagenic and carcinogenic effects (Mohammadi et al., 2022).

Acetaminophen (ACT), a widely used over-the-counter pain reliever, is commonly found in various concentrations in wastewater and natural water systems as a result of extensive consumption and disposal practices (Samal et al., 2022). Although often present in trace amounts, its persistence in the environment raises considerable concern. Like many pharmaceutical compounds, ACT resists biodegradation and tends to accumulate in aquatic ecosystems, posing risks not only to the environment but also to human and marine health (Yang et al., 2021).

Despite the absence of a globally standardized threshold for ACT concentrations in wastewater, continuous surveillance is crucial to reduce possible hazards to public health and ecological balance (Hai et al., 2018; Yang et al., 2021). This underscores the need for implementing advanced water treatment methods capable of effectively removing such contaminants. Among these, adsorption technologies have shown significant promise in purifying ACT-contaminated water, offering a viable means of maintaining safe water standards (Alrefaee et al., 2024; Snodin, 2023).

A wide range of methods have been established to eliminate ACT from polluted water sources, involving biological, photochemical, and oxidation-based treatments. However, adsorption occurred as a particularly efficient technique, favored for its affordability, operational simplicity, and strong performance even at low contaminant concentrations (Qasim et al., 2023). A wide range of adsorbents – from treated organic wastes to commercial activated carbon – has been investigated for their ability to capture ACT from aqueous environments (Khan et al., 2021). Despite extensive research over the years, a comprehensive and targeted review focusing specifically on ACT adsorption remains lacking. Given its extensive global usage, ACT continues to be a key pharmaceutical compound of interest in environmental remediation studies (Pratap et al., 2023).

Activated carbon is among the most extensively employed materials for removing contaminants from wastewater through adsorption. However, its use is frequently constrained by excessive production expenses and challenges in availability (Gulamhussein et al., 2023). This has intensified the search for alternative solutions that are not only efficient but also affordable and environmentally sustainable (Kulišťáková, 2023). In recent years, significant attention has been directed toward identifying low-cost adsorbents capable of eliminating pharmaceutical residues from water (Nguyen et al., 2021). Agricultural by-products, particularly fruit peels, have emerged as promising candidates (Ramos et al., 2023). Utilizing such materials offers a dual benefit – providing an economical alternative to traditional adsorbents and contributing to waste reduction, thus supporting environmental preservation (Ali et al., 2023; Skwarczynska-Wojsa and Puszkarewicz, 2024).

Nontraditional materials like rice husk, corn cob, banana peel, and coconut shell have all been studied for their ability to adsorb pharmaceutical compounds from water, demonstrating comparable performance in removing both organic and inorganic pollutants (Villaescusa et al., 2011). These alternatives are not only cost-effective but also present a greener option compared to activated carbon (Jedynak et al., 2019).

This study focuses on evaluating the potential of coconut shell waste and tree leaves as biosorbents for acetaminophen removal from aqueous solutions. Both materials were chemically modified using potassium hydroxide (KOH) and phosphoric acid (H_3PO_4) to enhance their adsorption properties. The impacts of key operational factors – pH, contact duration, initial ACT concentration, and the dosage of the adsorbent were meticulously analysed to ascertain ideal circumstances. Additionally, the adsorption behaviour of ACT on these biosorbents was examined utilising kinetic models, specifically pseudo-first as well as pseudo-second order, while equilibrium data were interpreted through Langmuir, Freundlich, and Temkin isotherm models to gain insight into the fundamental mechanics of adsorption.

MATERIALS AND METHODS

Adsorbent media

Tree leaves and coconut shell waste were chosen as economical agricultural by-products

for use in this study. Equal quantities of each material (500 grams) were naturally air-dried, then grounded utilized a food processor and passed through a standard mesh sieve (No. 100, 150 mm) to produce a fine powder. The produced powdered substances were kept in locked plastic containers at room temperature until they were needed again. Sodium hydroxide (NaOH), sulphuric acid (H_2SO_4), phosphoric acid, as well as potassium hydroxide included among the analytical-grade compounds used as experimental reagents. Acetaminophen, acting as the intended pollutant in the water-based solutions, was sourced from Aladdin Industrial Corporation, based in Shanghai, China. Table 1 presents the general properties of the acetaminophen used, which was utilized in its original form without any additional treatment.

Modification of biosorbents

Modification with KOH

To improve the existence of oxygen-rich functional groups on the adsorbent surface, the powdered material underwent chemical treatment using KOH. A total of 200 grams of the adsorbent was combined with 1000 mL of a 0.1 M potassium hydroxide solution and shaked at 200 rpm for three hours. After the treatment, the material was separated from the liquid and rinsed repeatedly using deionized water until the pH became neutral. The prepared adsorbent was then oven-dried at 105 °C and kept in a desiccator until further application (Kurniawan et al., 2011; Yanyan et al., 2018).

Oxidation of adsorbents with H_3PO_4

The adsorbent material underwent an oxidation process at room temperature by combining 200 grams of the sample with 200 mL of 65% phosphoric acid (H_3PO_4). The mixture was mixed at 100 rpm for three hours. After the reaction was finished, it was let cool down, thoroughly washed with deionized water, and then dried. The treated adsorbent was finally placed in a desiccator for storage until further use (Kurniawan et al., 2010; Yanyan et al., 2018).

Acetaminophen batch adsorption

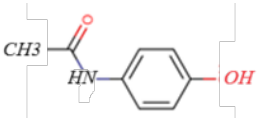
In the batch experiments, the required quantity of adsorbent was added to a determined concentration of ACT solution in a 100 mL beaker, adjusted to specific pH levels and temperatures. The mixture was then agitated at 200 rpm for varying time intervals. pH adjustments were made using different concentrations of sulfuric acid and sodium hydroxide solutions. At regular intervals, 5 mL Samples have been gathered for testing. To identify the optimum conditions for the adsorption process, variables such as adsorbent dose, reaction duration, and pH were altered one at a time while keeping other factors constant. Once equilibrium was reached, the solution was allowed to settle for at least 30 minutes. The supernatant was then collected from a point about 2 cm below the surface of the liquid, filtered through Whatman 542 filter paper, and analyzed. every experiment was carried out repeatedly multiple times, and the standard deviations was utilized to determine the standard error and variability of the data. Efficiency was established by the difference among the average initial and final concentrations of ACT. Random samples were periodically analyzed to verify and estimate the results.

The quantity of ACT adsorbed onto the adsorbent and the corresponding removal efficiency were calculated using the following expressions:

At time t (q_t), the adsorption capacity was measured by comparing the concentrations of the initial as well as final ACT levels, taking into account the adsorbent's mass and liquid volume:

$$q_t = \frac{(C_\circ - C_t)\, V_s}{w} \tag{1}$$

Table 1. General characterists of acetaminophen (Tao et al., 2015)

Chemical formula	λmax (nm)	Molecular weight (g/mol)	Solubility (g/L, 20 °C)	pKa	Molecular structure
$C_8H_9NO_2$	243	151.16	14	9.5	

The percentage of removal efficiency (R%) was calculated as the percentage reduction in ACT concentration relative to its initial value:

$$R\ (\%) = \frac{(C_\circ - C_t)}{C_\circ} \times 100 \quad (2)$$

where: V is the solution's volume in litres, W is the adsorbent's mass in grammes, R is the percent of ACT obtained from the solution, while q_t is the quantity of ACT adsorbed in mg/gram of adsorbent. The starting ACT concentration (in mg/L) is denoted as C_0, and the concentration at time t (in mg/L) is denoted as C_t.

Adsorption kinetic models

The adsorption process can be interpreted through various mathematical models that help in understanding the interaction mechanisms between adsorbates and adsorbents. These models are crucial for optimizing the design and operation of water and wastewater treatment systems. In this study, the kinetics of ACT adsorption were examined using two commonly applied models: the pseudo-first-order and pseudo-second-order models. The The pseudo-first-order model, which is stated as follows, suggests that the rate at which adsorption sites are occupied is proportionate to the number of empty sites:

$$Ln(q_e - q_t) = Ln\, q_e - k_1 t \quad (3)$$

The pseudo-second-order model, which suggests that the square of the quantity of accessible sites determines the adsorption rate, is as follows:

$$\frac{dq_t}{d_t} = k(q_e - q_1)^2 \quad (4)$$

where: q_t (mg/g) is the mass of ACT adsorbed at time t, q_e (mg/g) is the mass adsorbed at equilibrium, k_1 (1/min) is the pseudo-first-order rate constant, k_2 (g/mg·min) is the pseudo-second-order rate constant.

By integrating Equation 4 with initial conditions ($t = 0$ to $t = t$ and $q_t = 0$ to $q_t = q_t$), the model can be linearized as:

$$\frac{t}{q_t} = \frac{1}{k_2(q_e)^2} + \frac{t}{q_e} \quad (5)$$

This form allows for the evaluation of adsorption kinetics and helps identify A highly appropriate model for describing the data from experiments.

Adsorption isotherms

Adsorption isotherms are generally utilized to assess and interpret the behavior of adsorption processes. Among the most widely applied models for analyzing experimental isotherm data are the Langmuir, Freundlich, and Temkin models. These models are useful for describing the equilibrium state interaction between an adsorbent and an adsorbate. The corresponding equations for every model are summarized in Table 2.

Definitions of the parameters used in adsorption isotherm models are as follows:

- q: The quantity of material adsorbed (mg/g) per mass of adsorbent.
- q_m: Maximum adsorption capacity representing monolayer coverage (mg/g).
- n: Freundlich constant indicating the intensity of adsorption.
- C: At equilibrium, the concentration of the adsorbate in the solution is measured in milligrammes per litre.
- C_s: Solute concentration at saturation across all layers (mg/L).
- K_L: The Langmuir constant (L/mg) is correlated with binding site affinity.
- K_F: Freundlich constant representing adsorption capacity (mg/g).
- R: Universal gas constant, equal to 8.314 J/(mol·K).
- A_T: Temkin constant indicating equilibrium binding energy (L/g).
- b_T: Temkin constant related to heat of adsorption.
- q_s: Theoretical maximum adsorption capacity as per the isotherm model (mg/g).

Characterization of the modified adsorbents

To examine morphological alterations, dried chemically modified samples were mounted on a metallic stub using double-sided conductive adhesive tape. These samples were then gold-coated for 2 minutes using a sputter coater and analyzed under a Leica Stereo Scan 440 scanning electron microscope (SEM) equipped with an EDAX DX4i probe for energy dispersive X-ray (EDX) microanalysis. Due to the relatively large particle size, the materials were ground into a fine powder and dispersed in methanol prior to imaging. Surface functional group changes following chemical modification were assessed via Fourier transform infrared spectroscopy (FTIR) using a

Table 2. The adsorption isotherm models' mathematical formulas

Model	Original form	Linearized form
Langmuir	$q = \frac{q_m . K_L . C}{1 + K_L . C}$	$\frac{C}{q} = \frac{1}{q_m . K_L} + \frac{C}{q_m}$
Freundlich	$q = K_F . C^{\frac{1}{n}}$	$\log q = \log K_F + \frac{1}{n} \log C$
Temkin	$q = \frac{\mathbb{R}\, T}{b_\mathrm{T}} \ln(A_\mathrm{T} C)$	$q = \frac{\mathbb{R}\, T}{b_\mathrm{T}} \ln A_\mathrm{T} + \frac{\mathbb{R}\, T}{b_\mathrm{T}} \ln C$

Bruker Vektor 22 spectrometer under dry air at ambient conditions. About 6 mg of dried sample was mixed with 200 mg of potassium bromide (KBr), and from this blend, 40 mg was compressed into a pellet for analysis. The structural characteristics of the treated adsorbents were further investigated using X-ray powder diffraction (XRD) with a Philips Xpert system. The system operated with a copper anode and nickel filter, emitting radiation Cu Kα (λ = 1.54 Å) at 40 kV and 100 mA. Data on dispersion were gathered across a 2θ range of 5° to 100°, with readings taken every 0.05° and a count duration of 10 seconds per step. Each sample was mounted on a glass slide for measurement.

RESULTS AND DISCUSSION

Characteristics of adsorbent material

FTIR analysis was conducted within the spectral range of 400–4000 cm^{-1} to identify the surface functional groups on the adsorbents following chemical treatment. Figure 1 illustrates the FTIR spectra of both coconut shell waste and tree leaf-based adsorbents before and after modification (Repo et al., 2011). Broad absorption bands around 3400–3600 cm^{-1} reflect O–H stretching vibrations, indicating the presence of surface hydroxyl groups, carboxylic functionalities, and hydrogen-bonded water molecules (Tu et al., 2021). These peaks suggest strong hydrogen bonding interactions with water and functional groups. The O–H stretching was observed at 3414 cm^{-1} (AW), 3372 cm^{-1} (AW-H_3PO_4), 3339 cm^{-1} (AW-KOH), 3426 cm^{-1} (CS), 3420 cm^{-1} (CS-H_3PO_4), and 3414 cm^{-1} (CS-KOH), with reductions in peak intensity in some chemically modified samples, indicating partial dehydration during treatment. Aliphatic C–H vibrations were detected at 2928 and 2855 cm^{-1}, suggesting the presence of amino acids. Below 2000 cm^{-1}, characteristic peaks associated with structural and surface oxygen groups were identified. Carbonyl (C=O) stretching bands appeared near 1600–1630 cm^{-1} across all samples, slightly shifting based on the type of modification, with conjugation to aromatic structures influencing the peak positions (Abudi et al., 2025; Spataru et al., 2016). The broad bands around 1000 cm^{-1} were attributed to C–O ether groups, while absorptions near 600–620 cm^{-1} corresponded to hydroxyl ions. Peaks below 600 cm^{-1} were indicative of mixed metal oxides, particularly prominent in modified samples, appearing at 488cm^{-1} (AW-H_3PO_4), 555 cm^{-1} (AW-KOH), 604 cm^{-1} (CS), and 490 cm^{-1} (both CS-H_3PO_4 and CS-KOH). The FTIR analysis confirmed the presence of multiple oxygen-containing functional groups on the surface of the adsorbents, which are likely to play a crucial role in the adsorption of acetaminophen from aqueous solutions (Repo et al., 2009). In order to assess the surface morphology as well as elemental composition, energy-dispersive X-ray spectroscopy and scanning electron microscopy were employed, with magnification levels of 2 and 100 kX, respectively (Gomez-Serrano et al., 1996).

As shown in Figure 2, the SEM images revealed a rough and irregular surface with fragmented structures and dispersed grain sizes, some of which had formed agglomerates. These features included visible voids and pores, which are advantageous for enhancing surface area and, subsequently, adsorption efficiency (Nourmoradi et al., 2018; Su et al., 2021). The H_3PO_4-treated samples (AW-H_3PO_4 and CS-H_3PO_4) displayed a high density of cavities and a porous texture, with largely spherical particles. In contrast, the KOH-treated variants (AW-KOH and CS-KOH) presented smoother surfaces with interconnected microparticles (Shooto, 2023). These observations are consistent with earlier studies and suggest that chemical activation contributes to forming more uniform surfaces with finer pores, enhancing the material's adsorption potential. Elemental analysis using EDS further confirmed the elemental

makeup of the samples (Liu et al., 2023). As shown from Figure 3, the AW, AW-H_3PO_4, and AW-KOH materials contained elements like carbon, oxygen, magnesium, aluminum, silicon, potassium, and calcium, with additional components such as chlorine, phosphorus, sulfur, and iron detected in some variants, likely from additive residues (Ngernyen et al., 2023; Selvaraj et al., 2023). Coconut shell samples and their treated forms exhibited carbon, oxygen, phosphorus, and potassium, with aluminum present only in the H_3PO_4- and KOH-modified samples. Elements like magnesium, silicon, and calcium were also observed in CS-KOH. An increase in oxygen content across all adsorbents was evident, suggesting the incorporation of oxygen-containing groups due to chemical treatment, likely through sulfonation processes (Aliyu et al., 2022; Tran et al., 2020).

The findings of the XRD study of the adsorbent composites are illustrated in Figure 4, showing details about the materials' structural properties. XRD was employed to distinguish between crystalline and amorphous phases – sharp, well-defined peaks indicate crystallinity, while broad, diffuse peaks suggest an amorphous nature (Najafi et al., 2023; Wakejo et al., 2023). The patterns for both AW and CS revealed prominent crystalline cellulose peaks, observed at 2θ values of 15.10° and 22.65°, respectively. In the AW sample, pronounced reflections at 2θ = 20.85° and 23.35° were likely associated with graphite structures. Additionally, a peak near 2θ = 26.9° signaled the presence of silicon oxide. Peaks appearing at 2θ = 30.15° and 36.15° in AW, and at 35.15° in CS, further indicated silicon oxide and calcium-based compounds. A reflection at 2θ = 38.25° corresponded to calcium oxide (Arif et al., 2023; Zubair et al., 2023). Both AW and CS displayed broad peaks at higher angles (2θ = 61.50° and 62.25°), which were linked to the (214) planes of graphitic carbon. The AW-KOH composite exhibited a distinct and intense peak at 2θ = 11.50°, suggesting successful structural modification through potassium hydroxide treatment. However, the XRD profile of CS-KOH closely resembled that of untreated CS, implying minimal structural change from the modification, despite some anticipated effects due to potassium addition (Wen et al., 2022). For the H_3PO_4-treated samples (AW-H_3PO_4 and CS-H_3PO_4), the XRD patterns lacked sharp peaks, reflecting a transformation towards an amorphous structure. This is due to phosphoric acid's capacity for promotion the formation of amorphous carbon by disrupting the crystalline framework (Chang et al., 2021; Z. Zhang et al., 2022). The data also highlighted the high structural purity of the KOH-modified adsorbent, as evidenced by the absence of extraneous diffraction peaks (Hamid et al., 2022; H. Zhang et al., 2022). Conversely, the changes introduced by H_3PO_4 modification were more substantial, as seen in the significant alterations in the XRD patterns, pointing to a marked reorganization of the material's internal structure (Al-Shehri et al., 2023).

Parameters affecting ACT adsorption

Solution pH Impact

Effects of pH on ACT removal from water using various adsorption materials (AW, AW-H_3PO_4, AW-KOH, CS, CS-H_3PO_4, and CS-KOH) were examined (Figure 5). The optimal pH for AW and CS was found to be 6, with removal efficiencies of 46.82% and 49.28%, respectively (Villaroel et al., 2014). For AW-KOH, CS-KOH, and CS-H_3PO_4, the optimal pH was 8, with removal efficiencies of 82.53%, 88.86%, and 85.01%, respectively. AW-H_3PO_4 showed the best removal efficiency (78.97%) at pH 4. As shown in Figure 5, ACT adsorption increased initially from pH 2.0 to 6.0, reaching maximum removal rates of 46.82%, 49.28%, and 78.97% for AW, CS, and AW-H_3PO_4, respectively, at pH 6.0. [63] However, AW-H_3PO_4 reached maximum adsorption earlier at pH 4. This finding aligns with Villaroel et al. (2014) (Villaroel et al., 2014), who noted higher ACT deterioration in acidic conditions (pH 3.0–5.6) compared to basic solutions (pH 9.5–12.0). For AW-KOH, CS-KOH, and CS-H_3PO_4, the optimum pH was 8, with removal efficiencies of 82.53%, 88.86%, and 85.01%, respectively. An additional rise in pH beyond 8 resulted in a notable decline in ACT removal. The varying responses of the adsorbents to different pH values suggest that different adsorption mechanisms are involved, with electrostatic attraction playing a key role in the process. The improved removal effectiveness of altered adsorbents highlight the positive impact of modification. The pH effect on adsorption can be attributed to ACT's weak electrolyte nature, existing in ionized or nonionized forms depending on the solution's pH relative to its pKa value (9.38) (Igwegbe et al., 2021) (Wakejo et al., 2023). When the pH is below ACT's pKa, it primarily exists in a neutral,

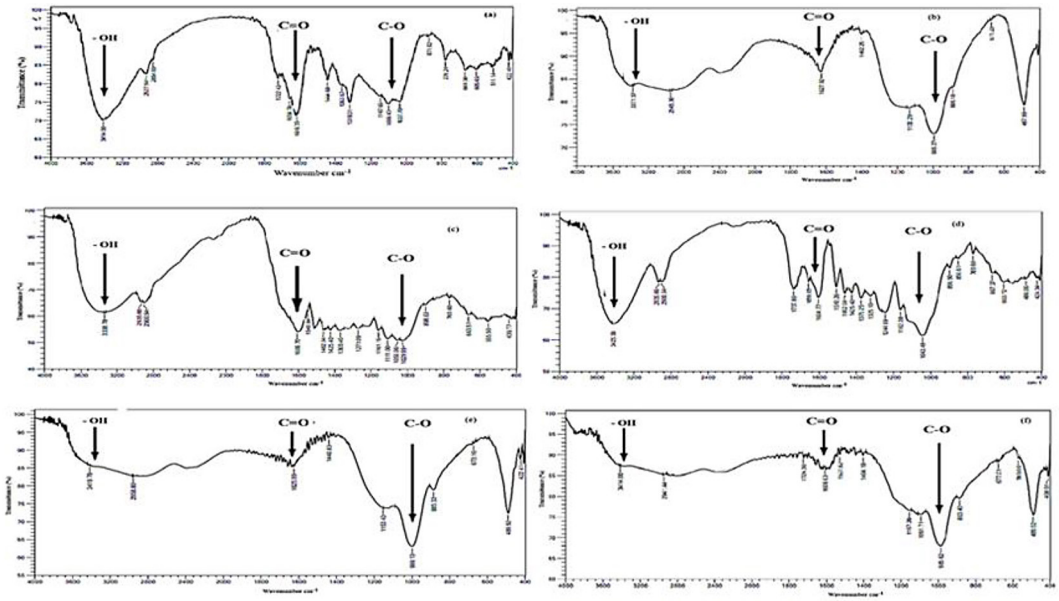


Figure 1. FTIR spectra of the adsorbent materials following surface modification with various chemical agents, including (a) AW, (b) AW-H_3PO_4, (c) AW-KOH, (d) CS, (e) CS-H_3PO_4, and (f) CS-KOH.

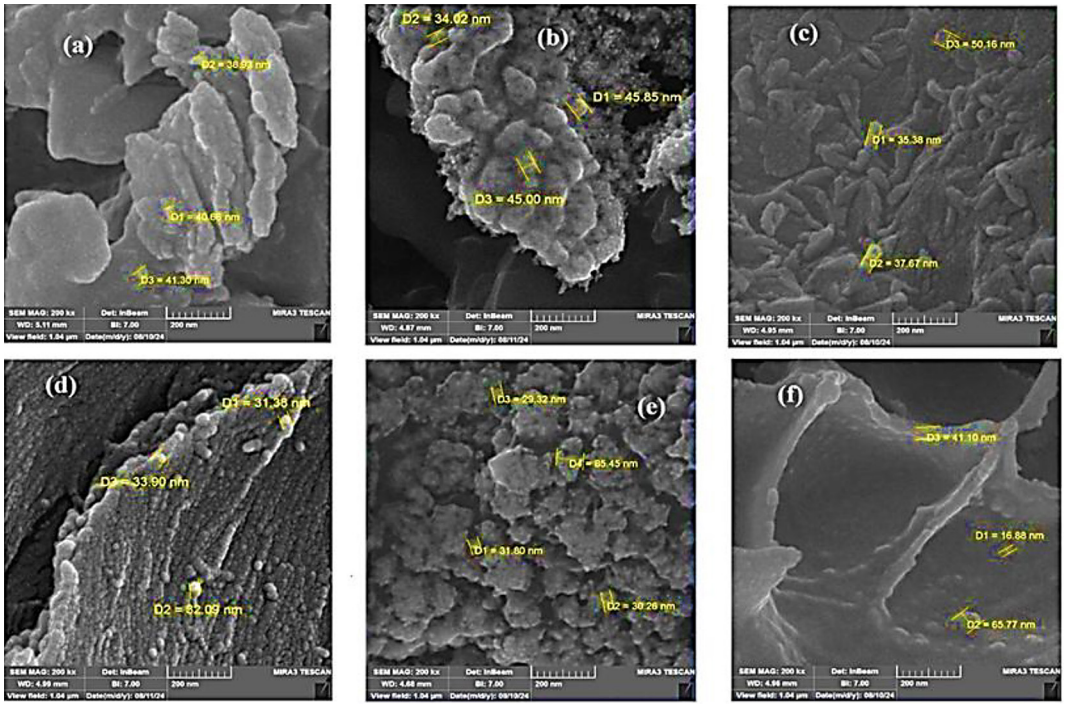


Figure 2. SEM images of the following samples: (a) AW, (b) AW-H_3PO_4, (c) AW-KOH, (d) CS, (e) CS-H_3PO_4, and (f) CS-KOH.

nonionized form, reducing electrostatic attraction and thus impacting adsorption efficiency (Abudi, Al-Saedi, et al., 2025).

Effect of adsorbent dose

The impact of adsorbent dose on the removal of ACT was evaluated by varying the dose from 0.1 to 1.1 g/L and adding it to an 80 mg/L ACT solution for 150 minutes at the optimal pH for each adsorbent. As shown in Figure 6, the adsorption efficiency improved with an increase in the adsorbent dose. For both AW, CS, and their modified forms, removal efficiencies rose from 5.41% to 77.99% for AW, 38.10% to 93.26% for AW-KOH, 38.76%

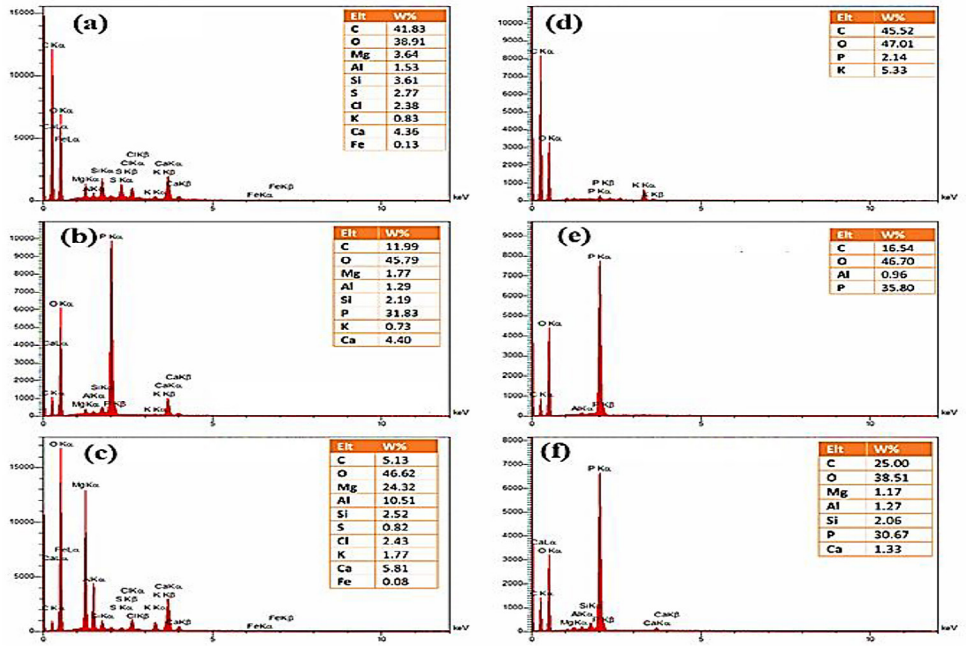


Figure 3. EDS analysis of the adsorbents and their modifications: (a) AW, (b) AW-H_3PO_4, (c) AW-KOH, (d) CS, (e) CS-H_3PO_4, and (f) CS-KOH.

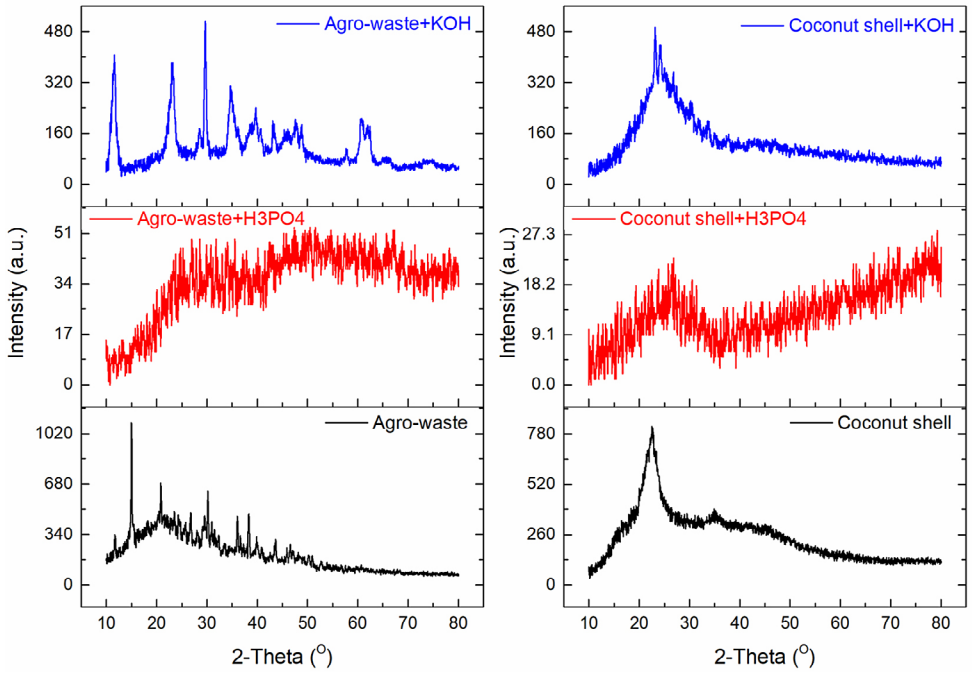


Figure 4. XRD patterns for biosorbents

to 93.47% for AW-H_3PO_4, 13.38% to 76.60% for CS-KOH, 44.99% to 93.78% for CS-H_3PO_4, and 39.10% to 93.64% for CS, as the dose increased from 0.10 to 1.1 g/L. This increase can be attributed to the additional sorptive surface area available with a higher adsorbent mass, resulting in more active sites for adsorption (Wakejo et al., 2023) (Abudi, Hameed, et al., 2025). Raising the dosage from 0.9 to 1.1 g did not significantly enhance removal efficiency, though, suggesting that higher doses beyond 0.9 g are unnecessary. Additionally, at higher doses (0.9 and 1.1 g), the modified adsorbents performed significantly better than the raw materials. Similar studies conducted by other

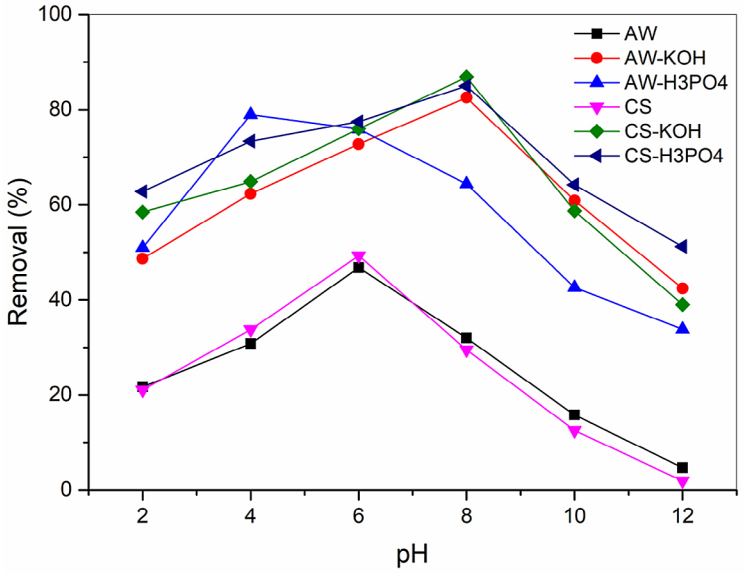


Figure 5. pH Impact on acetaminophen elimination efficiency (with initial concentration of 80 mg/L, dose of biosorbent = 0.3 g, contact time = 150 min)

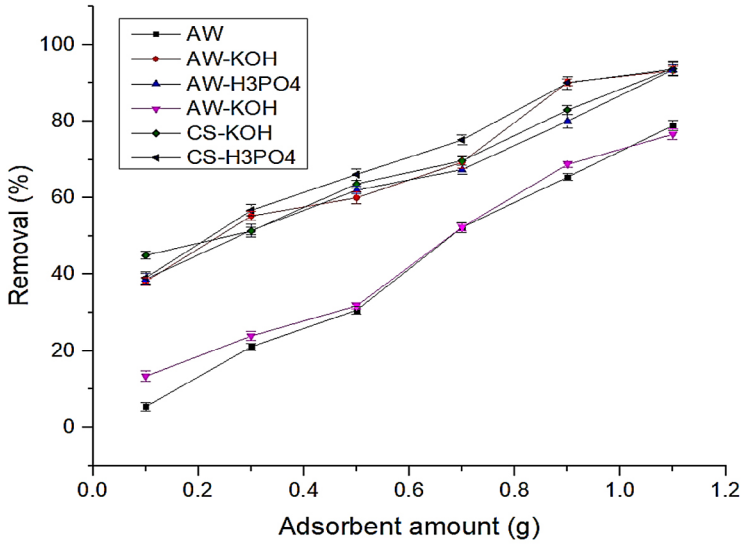


Figure 6. Adsorbent dose influence on acetaminophen removing efficiency

researchers also explored the effect of various adsorbent doses under different conditions. To treat a sample containing 2000 mg/L of acetaminophen, for instance, Manee Wong et al. (2022) used 0.08 g/100 ml of activated carbon from coconut shell (Maneewong et al., 2022), in contrast to that, dos Reis et al. (2022) used tree bark waste at a concentration of 0.15 g/100 ml to remove acetaminophen from water solutions containing 70–1200 mg/L.

Effect of acetaminophen initial concentration

The adsorption of acetaminophen by AW, CS, and their modified forms was studied, as the initial concentration of the adsorbate plays a crucial role in the adsorption process. The initial concentration serves as an important driving force to overcome the mass transfer resistance between the aqueous and solid phases (Abudi et al., 2025). Figure 7 illustrates the effect of the initial ACT concentration within the range of 40–200 mg/L, using 1.1 g of adsorbent for 150 minutes at the optimal pH for each material. The removal efficiency increased from 86% to over 93% when the ACT concentration rose from 40 to 80 mg/L. However, when the concentration exceeded 80 mg/L, the removing efficiency

dropped significantly, reaching as low as 31%. This reduction in efficiency with higher ACT concentrations could be related to the limited availability of adsorption sites and a reduction in intraparticle diffusion (Liu et al., 2023). In this study, the maximum equilibrium uptake (qe) was reached at 120 mg/L, with values of 7.09 and 6.82 mg/g for AW and CS, respectively. The modification of adsorbents notably enhanced the qe, with values of 8.34, 8.13, 8.16, and 8.71 mg/g for AW-KOH, AW-H_3PO_4, CS-KOH, and CS-H_3PO_4, respectively, at a concentration of 150 mg/L. Beyond 150 mg/L, the removal efficiencies dropped sharply as the adsorbents reached their maximum capacity for uptake. However, the equilibrium uptake (qe) values in this study were significantly lower compared to those found in other studies. For example, dos Reis et al. (2022) found values surpassing 200 mg/g for a tree bark-based adsorbent, whereas Maneewong et al. (2022) observed qe values close to 200 mg/g for biochar derived from coconut shells (Maneewong et al., 2022). This substantial difference can likely be attributed to variations in the characteristics of the raw materials and the experimental conditions used.

Effect of contact time

The impact of contact time on ACT adsorption was investigated over a period ranging from

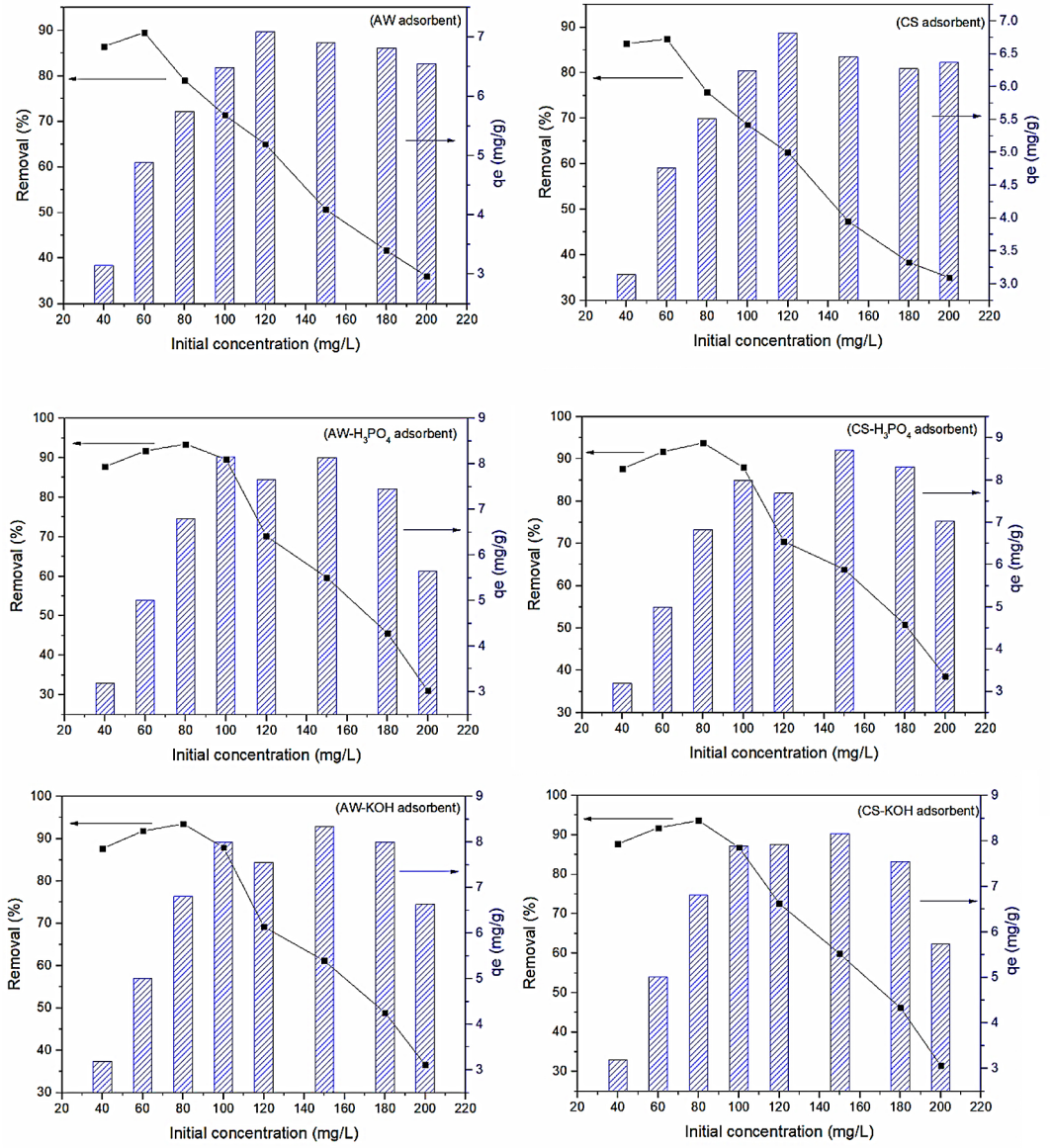


Figure 7. Impact of initial acetaminophen concentration on adsorption at equilibrium (qe) and removal efficiency

5 to 150 minutes at an ACT concentration of 120 mg/L and an adsorbent dose of 1.1 g/L, with each adsorbent at its optimum pH (Loc et al., 2023). As shown in Figure 8, the effectiveness of removal raised dramatically with contact time, showing that more than 120 minutes are required to reach removal efficiencies above 90%. The modification of AW and CS notably improved the removal rate, especially after 120 minutes. Initially, the presence of free pores facilitated the adsorption process, but extending the contact time beyond 120 minutes did not lead to substantial increases in ACT removal, as equilibrium was reached. This trend aligns with findings by Natarajan et al. (2021). While slower adsorption indicates that pore occupancy is the primary process, fast adsorption is usually linked to surface adsorption and an increased reactivity between acetaminophen molecules and the adsorbent (Loc et al., 2023).

Kinetic and isotherm studies

Utilizing kinetic variables, the adsorption process was quantitatively analyzed employing pseudo-first and pseudo-second order kinetic models (Table 3). The R^2 values for the pseudo-second order model were generally higher (0.96, 0.89, 0.91, 0.95, 0.90, and 0.91) compared to those for the pseudo-first order model (0.89, 0.87, 0.85, 0.93, 0.87, and 0.90) for AW, AW-KOH, AW-H_3PO_4, CS, CS-KOH, and CS-H_3PO_4, respectively, providing more accurate descriptions of the kinetic data than the pseudo-second order model. The adsorption process was better portrayed by this model, which posits that chemisorption is the main mechanism and that the adsorption capacity is highly dependent on the availability of active sites on the adsorbent surface. In line with the results reported by Lung et al. (2021), Table 3 shows that the adsorption rate was slower due to the comparatively low values of K2. On the other hand, adsorbents derived from tree bark were found to have a quicker mechanism for acetaminophen adsorption in the study by dos Reis et al. 2022.

The Langmuir, Freundlich, and Tempkin models were used to examine the adsorption isotherms, with the corresponding parameters presented in Table 4. The R^2 values for the Langmuir model (0.97, 0.97, 0.91, 0.98, 0.93, and 0.97 for AW, AW-KOH, AW-H_3PO_4, CS, CS-KOH, and CS-H_3PO_4, respectively) stood out from the Freundlich as well as Tempkin models, suggesting that the Langmuir model more precisely portrayed the adsorption process and offered a more appropriate fit for the isothermal data. The results are in agreement with those of the study done in 2021 by Kerkhoff et al. (2021). The Langmuir model states that when the adsorbent surface is uniform along with the adsorbed molecules are not in contact with each other, single-layer adsorption takes place (Quesada et al., 2019). In contrast, the Freundlich model revealed a weaker correlation with the experimental data (R^2 values of 0.91, 0.68, 0.80, 0.80, 0.80, and 0.87 for AW, AW-KOH, AW-H_3PO_4, CS, CS-KOH, and CS-H_3PO_4, respectively). The Freundlich model,

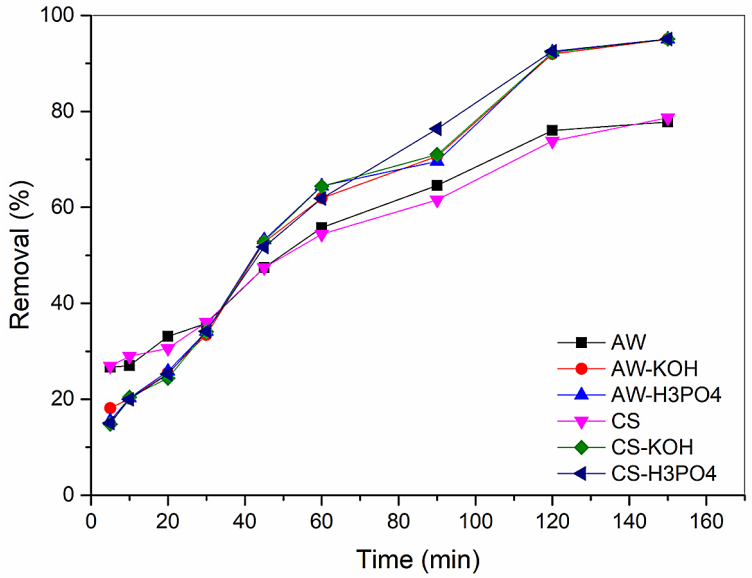


Figure 8. Impact of contact time on acetaminophen removal

Table 3. Kinetics study parameters

Adsorbent	Parameter		Pseudo-first order			Pseudo-second order		
	pH	qe, exp (mg/g)	k1 (1/min)	qe, cal. (mg/g)	R^2	k2 (g/mg/min)	qe, cal. (mg/g)	R^2
AW	6	8.4848	0.026	8.734	0.89	0.003	9.980	0.96
AW-KOH	6	8.6409	0.025	10.451	0.87	0.001	12.092	0.89
AW-H_3PO_4	6	8.6434	0.026	10.664	0.85	0.001	12.165	0.91
CS	6	8.5805	0.019	7.463	0.93	0.003	9.872	0.95
CS-KOH	6	8.6434	0.026	10.715	0.87	0.001	12.422	0.90
CS-H_3PO_4	6	8.6434	0.027	11.105	0.90	0.001	12.626	0.91

Table 4. Isotherm study parameters

Adsorbent	Langmuir isotherm				Freundlich isotherm			Tempkin isotherm		
	KL (L/mg)	qm, cal (mg/g)	R^2	RL	KF	n	R^2	KT	B	R^2
AW	0.349	5.102	0.97	0.182	5.004	21.097	0.91	0.000	-1.349	0.66
AW-KOH	0.407	9.699	0.97	0.165	5.635	7.380	0.68	117.294	1.096	0.67
AW-H_3P_O4	0.227	10.070	0.91	0.234	1.069	1.427	0.80	0.153	6.885	0.75
CS	0.224	4.831	0.98	0.236	13.095	-4.359	0.80	0.000	-1.282	0.81
CS-KOH	0.228	10.549	0.93	0.234	1.552	1.770	0.80	0.232	5.640	0.76
CS-H_3PO_4	0.257	11.416	0.97	0.219	4.856	4.572	0.87	7.930	1.859	0.85

which assumes multilayer adsorption and surface heterogeneity, still provided a reasonable fit, as reflected in the relatively moderate R^2 values.

CONCLUSIONS

The adsorption process effectively removed acetaminophen from aqueous solutions, with modifications to the raw adsorbents, such as agricultural waste and coconut shell, significantly improving the process. Key parameters such as initial ACT concentration, pH, contact time, and adsorbent dose were optimized using a one-factor-at-a-time approach. The optimal conditions identified were an adsorbent dose of 1.1 g/L, pH 8.0 for all modified adsorbents (except AW-H_3PO_4, which was optimized at pH 4), a contact time of 120–150 minutes, and ACT concentrations of 100–120 mg/L. Under these conditions, the modified adsorbents (AW and CS) achieved more than 95% removal of ACT, compared to around 78% with the raw material. The study demonstrated that the modified agricultural waste and coconut shell adsorbents, under optimized conditions, could remove acetaminophen with efficiencies of 78%, 95%, 95%, 95%, 79%, 95%, and 95% for AW, AW-H_3PO_4, AW-KOH, CS, CS-H_3PO_4, and CS-KOH, respectively. A substantial increase in the adsorbents' modified surface area, as compared to the raw materials, is associated with this enhancement. The equilibrium uptake of the modified adsorbents increased 1.10 to 1.35 times compared to the raw adsorbents. According to FTIR measurement, the adsorbents' chemical activation added advantageous surface functional groups for efficient ACT adsorption. KOH and H3PO4 activations both helped to increase the number of mesopores with lower pore diameters, which improved the adsorption process. The main components of all adsorbent combinations were carbon and oxygen, and the adsorbent's affinity for acetaminophen was enhanced by the inclusion of layer double hydroxides. The pseudo-second-order model was determined to fit the sorption data the best after kinetic and isotherm models were used. ACT adsorption onto the adsorbent surface took place in a monolayer on a heterogeneous surface, according to the isotherm analysis. In contrast to raw AW's 5.102 mg/g, the highest Langmuir sorption capacities for AW-H_3PO_4 and AW-KOH were 10.07 mg/g and 9.699 mg/g, respectively. The capacities for CS were 4.831 mg/g for raw CS and 11.416 mg/g for CS-H_3PO_4 and 10.549 mg/g for CS-KOH. Overall, acetaminophen extraction from aqueous solutions showed promising results using coconut shell and agricultural waste as eco-friendly adsorbents. These raw

adsorbents' capacity to remove acetaminophen was greatly increased by chemical modification, as shown by improvements in their properties. Additional investigation is required to examine the potential of these adsorbents for removing other pharmaceutical contaminants.

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