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Study of the Use of Activated Carbon Prepared from the Eichhornia Crassipes Plant for Removing Paracetamol from Aqueous Solutions

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Adsorption; Activated carbon; Eichhornia Crassipes; Kinetic models; Pharmaceutical contaminants.

Highlights:

- *Preparation of activated carbon from agricultural waste.*
- *Removal of pharmaceutical contaminants from aqueous solutions.*
- *Study of the properties of activated carbon prepared from Eichhornia crassipes.*

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Abstract: This study utilized activated carbon derived from Eichhornia crassipes (EC), a cheap plant that grows widely along riverbanks. The plant has fibrous roots and a taproot. It has thin, round, erect, wavy, smooth, and shiny leaves. The flowers are beautiful violet with six petals, and the fruits contain about 300 seeds each. The seeds can remain active for about 20 years. It negative impacts contain environment because it consumes large amounts of water, as a biosorbent in batch adsorption experiments for the removal of paracetamol from synthetic aqueous solutions. It was tested as an adsorbent for heavy metals, dyes, and other pollutants. The biosorbent was characterized using XRD, FTIR, BET, and SEM techniques. Several factors affecting the adsorption were considered in the study, including the adsorbent dosage (AC/NOH) (0.2-2.5 g/L), pH value (3-10), initial paracetamol concentration (10-80 mg/L), and contact duration (15-240 min). The highest removal efficiency of 75% for paracetamol was observed at a contact time of 180 min, pH value of 3, adsorbent dosage of 2 g, and initial paracetamol concentration of 10 mg/L at 25 °C, and the maximum adsorption capacity of 17.42 mg/g was observed with an R^2 value of 0.9851 on the Langmuir curve, while the pseudo-second order kinetic model provided a better fit with $R^2 = 0.9987$. The study utilizes Eichhornia Crassipes as a low-cost and environmentally friendly bio sorbent, which has not been explored in previous research on removing pharmaceutical contaminants. This result suggests its potential application in removing other pharmaceutical contaminants from aqueous solutions.

دراسة استخدام الكربون المنشط المُحضر من نبات إيكورنيا كراسيبس لإزالة الباراسيتامول من المحاليل المائية

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الخلاصة
استخدمت هذه الدراسة الكربون المنشط المستخلص من نبات زهرة النيل (Eichhornia crassipes)، وهو نبات رخيص الثمن ينتشر بكثرة على ضفاف الأنهار. يتميز هذا النبات بجذوره الليفية وجذره الوتدي، وأوراقه الرقيقة المستديرة المنتصبة المتموجة الملساء اللامعة. أزهاره بنفسجية جميلة بست بتلات، وتحتوي ثماره على حوالي ٣٠٠ بذرة. يمكن أن تبقى البذور قابلة للتحلل لمدة ٢٠ عامًا تقريبًا. على الرغم من تأثيره السلبي على البيئة نظرًا لاستهلاكه كميات كبيرة من الماء، فقد استُخدم الكربون المنشط كمادة ماصة حيوية في تجارب الامتزاز الدفعي لإزالة الباراسيتامول من المحاليل المائية الاصطناعية. كما تم اختباره كمادة ماصة للمعادن الثقيلة والأصباغ والملوثات الأخرى. وتم توصيف المادة الماصة الحيوية باستخدام تقنيات حيود الأشعة السينية (XRD) ومطيافية الأشعة تحت الحمراء بتحويل فورييه (FTIR) وقياس مساحة السطح النوعية (BET) والمجهر الإلكتروني الماسح (SEM). أخذت في الدراسة عدة عوامل مؤثرة على الامتزاز، بما في ذلك جرعة المادة المازة (AC/NOH) (٠,٢-٢,٥ غ/لتر)، وقيمة الرقم الهيدروجيني (٣-١٠)، والتركيز الأولي للباراسيتامول (١٠-٨٠ ملغ/لتر)، ومدة التلامس (١٥-٢٤٠ دقيقة). وقد لوحظت أعلى كفاءة إزالة للباراسيتامول بنسبة ٧٥٪ عند مدة تلامس ١٨٠ دقيقة، وقيمة رقم هيدروجيني ٣، وجرعة مادة مازة ٢ غ، وتركيز أولي للباراسيتامول ١٠ ملغ/لتر عند درجة حرارة ٢٥ درجة مئوية. كما لوحظت أعلى سعة امتزاز بلغت ١٧,٤٢ ملغ/غ بقيمة R^2 تساوي ٠,٩٨٥١ على منحنى لانغموير، بينما قدم نموذج الحركية من الرتبة الثانية الزائفة توافقًا أفضل بقيمة R^2 تساوي ٠,٩٩٨٧. تستخدم هذه الدراسة نبات إيكورنيا كراسيبس كمادة ماصة حيوية منخفضة التكلفة وصديقة للبيئة، وهو ما لم يُستكشف في الأبحاث السابقة المتعلقة بإزالة الملوثات الصيدلانية. تشير هذه النتيجة إلى إمكانية استخدامه في إزالة ملوثات صيدلانية أخرى من المحاليل المائية.

الكلمات الدالة: الامتزاز، الكربون المنشط، نبات إيكورنيا كراسيبس، النماذج الحركية، الملوثات الصيدلانية.

1.INTRODUCTION

Wastewater and industrial effluents pollute surface water, groundwater, and soil, causing environmental problems that harm both human health and aquatic ecosystems. Pharmaceutical waste, including paracetamol, is one of the most important pollutants discharged with wastewater, altering the chemical composition of water by changing its pH value, conductivity, and other chemical properties [1-3]. Pharmaceutical pollutants are classified as hazardous substances that can enter the environment through different sources, e.g., drugs, hospital waste, pharmaceutical industry, and harm the natural ecosystem and change the equilibrium [4, 5]. Analgesics, including paracetamol, are among the most widely consumed drugs worldwide. It is widely used as a pain reliever, antipyretic, and a major component of most influenza medications. It can be used without a prescription, and its consumption has increased since the COVID-19 pandemic. It has been observed that it is present in large proportions in wastewater and is not metabolized, posing a risk to both humans and aquatic life. Paracetamol was used as a model for our study. Recent studies have focused on improving water treatment technologies and developing effective methods to eliminate pharmaceutical micropollutants from contaminated water [6]. Several technologies have been proposed for treating organically contaminated wastewater, including electrochemical methods, sedimentation, coagulation and flocculation, membrane separation [7], advanced oxidation [8], and adsorption [9]. Adsorption is one of the most exciting technologies at present due to its efficiency, low cost, low toxicity, environmental friendliness, and effectiveness in eliminating pharmaceutical pollutants [10], activated carbon [11], multi-walled carbon nanotubes [9], waste tires [12], and low-cost sorbents [13;]14]. They have been used in the literature review to remove and adsorb harmful pollutants effectively and are environmentally friendly. Several studies have investigated the removal of paracetamol from aqueous solutions using advanced activated carbon prepared from various plant sources, including Cannabis sativa [15], Cordia myxa [16], Oak [17], and Orange peels [18]. In this research, Eichhornia Crassipes, is a prolific aquatic plant found on river margins. It is sustainable and widely available due to its ability to absorb non-conventional pollutants from water, such as colors and heavy metals. It has been used as a biosorbent in both natural and carbonated form in water treatment systems to produce purified water. It is a promising biosorbent [19]. The goal of this study is to develop an adsorbent from the Eichhornia Crassipes plant that effectively extracts paracetamol from synthetic aqueous solutions. To achieve this goal, the following steps were taken: (1) By producing activated carbon from Eichhornia Crassipes plants and treating them with potassium hydroxide (KOH), an environmentally conscious approach is promoted by reducing waste generation and disposing of the plant, which is detrimental to the aquatic environment due to its high-water consumption. (2) Examination of the activated carbon's characteristics using various characterization techniques. (3) Analyzing the efficiency of activated carbon in eliminating paracetamol from synthetic aqueous solutions under various operating circumstances, such as temperature, pH, stirring speed, AC/NAOH dosage, and paracetamol concentration. (4) Using the best kinetic, thermodynamic, and isothermal models and selecting them.

2. EXPERIMENTAL WORK

2.1.Materials

Hydrochloric acid (HCL) 37 extra pure (36.5 g/mol), Sodium hydroxide NaOH pure 99% (40g/mol), Potassium hydroxide (KOH) pure 99% (56 g/mol) were supplied by the AGFA Company of Berlin and purchased from Alhekma market. High-purity Paracetamol ($C_8H_9NO_2$) was obtained from the Samarra Pharmaceutical Factory in Iraq. Also, distilled water was used in the laboratory experiments.

2.2.Preparation of Paracetamol Synthetic Aqueous Solution

Paracetamol is abbreviated as (4-hydroxyacetanilide). It resembles a white, crystalline substance in form. Mwt is 151.165 g /mol, and the chemical formula is $C_8H_9NO_2$. It is utilized as a pain reliever and antipyretic in cases of fever [20]. Figure 1 shows the chemical structure of paracetamol. It was used to create a standard stock solution, with a concentration of 1000 mg/L by dissolving one gram of the medication in one liter of distilled water. All laboratory experiments were conducted in the Department of Environmental Engineering/ College of Engineering/ Tikrit University. The paracetamol concentrations were measured during the experiment using an ultraviolet spectrophotometer with a maximum wavelength of 243 nm through a series of dilutions that were conducted on the standard stock solution to obtain the required concentrations ranging from (5-80) mg/L [21].

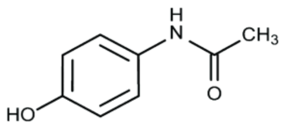


Fig. 1 Chemical Structure of Paracetamol [15].

2.3.Preparation of Activated Carbon from the Eichhornia Crasspies Plant

Eichhornia Crassipes was collected from the banks of the Tigris River in Al-Alam District, Salah al-Din Governorate, Iraq. To facilitate washing and drying, the plant parts were divided into stems, leaves, and roots. After washing it with tap water to remove contaminants, the plant was sun-dried for 5 days before being crushed and sieved through a 425 µm sieve. Then, it was washed with distilled water and thoroughly dried in an oven at 105 °C for 2 hours [22]. Activated carbon was prepared by thermal decomposition of carbon materials. A sample of the ground plant was placed in a pottery jar in a tubular burning oven at 600 °C for 2 hours, in the presence of inert nitrogen gas at a continuous flow rate of 100 ml/min prevent the material from turning to ash. After the resulting carbon was formed, it was rinsed with distilled water through a 0.45 µm filter paper to remove any contaminants or ash produced during the burning process. The raw carbon was then combined with the 0.5 N KOH solution at a 1:2 (w/v) impregnation ratio and left for 24 hours. It was then rinsed for 1 h with hot distilled water until the pH of the solution reached 7. The resulting carbon was then dried, making it ready for use in experiments, as shown in Fig. 2.

2.4.Characterization of Activated Carbon

Several tests were conducted on (AC/KOH), and an analyzer of surface area type, BELSORP, Microtrac Co, Japan, was employed to calculate the surface area (Brunauer-Emmett-Teller (BET) (BELSORPMINI I, Japan). The functional groups of the carbon surface were studied using FTIR spectroscopy and a spectrometer (Shimadzu, Japan). XRD, X-Ray photoelectron spectroscopy (PW1730, Philips, Netherlands), was applied to investigate the samples. Physical characterization of the activated carbon was investigated using a SEM, Scanning Electron Microscope, (JSM-6060 LV, USA).

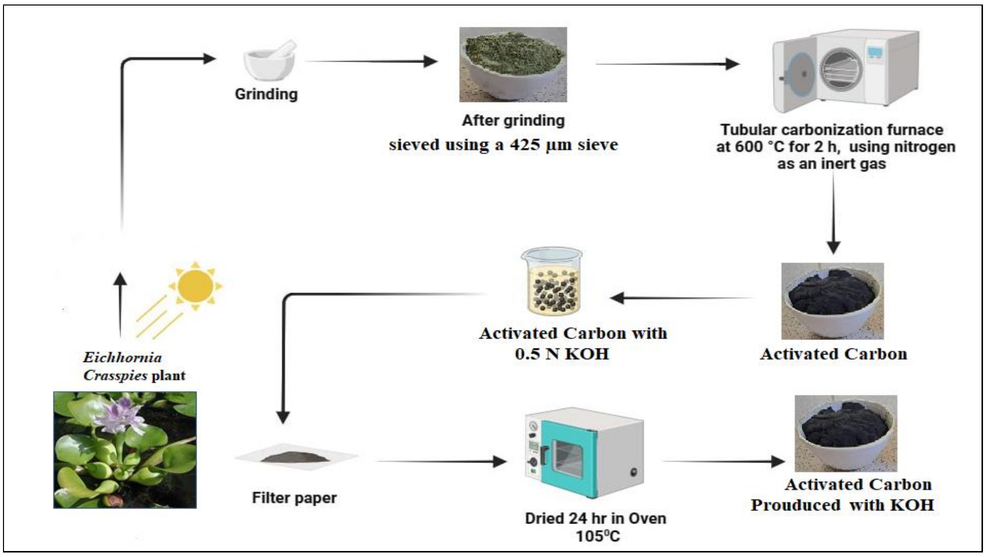


Fig. 2 Steps Preparation of Activated Carbon from the Eichhornia Crassipes plant and Activation with KOH.

3.ADSORPTION EXPERIMENTS

The optimum parameters for the removal of paracetamol at room temperature were calculated utilizing batch adsorption experiments, at different operating conditions, including pH (3,4,6.5,7, 9, and 10), initial paracetamol concentration (10,30,40,60,and 80 mg/L), contact time (15 to 240 min), AC/KOH dosage (0.010, 0.020, 0.030,0.040,0.050,0.075,0.100, and 0.150g/50 mL), and shaking speed (100,150,and 200 rpm). In a 100 ml glass beaker, 50 ml of the aqueous solution of paracetamol was added, and a specific amount of the absorbent material was added at the pH value determined. The aqueous solution was continuously shaken at 150 rpm and 25 ± 2 °C using a mechanical shaker, type digital orbital shaker, (SHO_2D, England). The pH was adjusted using 0.1 M HCl or 0.1 M NaOH, until the pH values were as required, using a pH instrument (Hana 211, Romania). Samples are filtered on a 42 µm filter paper to separate the activated carbon aqueous solution. To calculate the paracetamol removed in the supernatant, a spectrophotometric and double-beam spectrophotometer type Single Beam, Spectro UV-2550, Norway, were used to determine the removal of paracetamol absorbance at a wavelength of 243 nm. Eqs. (1) and (2) are used to determine the capacity of adsorption (*q*) and removal effectiveness (R%) (Huang et al., 2014):

$$q_e = \frac{(C_\circ - C_e)\,V}{m} \qquad (1)$$

where q_e is the amount of adsorbent per unit gram of adsorbent (mg/g), C_o is the adsorbent's initial concentration, C_e is the adsorbent's equilibrium concentration (mg/l), m is the adsorbent's mass (g), and V is the solution's volume (L). The effectiveness of removing the adsorbent from the adsorbent at Time (t) is obtained by the following equation:

$$R\% = \frac{(C_\circ - C)}{C_\circ} \qquad (2)$$

where R = is the percentage of removal efficiency.

4.MODELS OF ISOTHERM

Isotherm Freundlich, Langmuir, Temkin, and Dubinin-Radushkevich models are used to calculate the equilibrium between sorbate and adsorbent, i.e., the ratio at a constant temperature and the balance between what is absorbed and what remains in solution [23]. Also, these models provide a framework for studying and forecasting adsorption behavior in a wide range of scientific and industrial applications, including catalysis, environmental remediation, and material development. The applicability of each model is determined by the specific properties of the adsorption system under study.

Langmuir isothermal model [24] is represented in the following equation:

$$q_e = \frac{q_m K_l C_e}{1 + K_l C_e} \qquad (3)$$

where q_m is the adsorption capacity, K_l is the Langmuir constant, and q_m is the maximum adsorption capacity. The nature of adsorption is shown by the relationship between the adsorbent and adsorbate, as in the following equation:

$$R_L = \frac{1}{1 + K_1 C_o} \qquad (4)$$

The value of R_L represents four distinct isotherm situations: unfavorable ($R_L > 1$), linear ($R_L = 1$), or advantageous ($R_L < 1$), and irreversible ($R_L = 0$) [9]. The Freundlich isothermal model determines the heterogeneous nature of the adsorbed surface and the corresponding energy, indicating the maximum adsorption capacity [25]. The equation is expressed as follows:

$$q_e = K_f C_\circ^{\frac{1}{n}} \qquad (5)$$

where K_f is the adsorption capacity, and 1/n is the adsorption intensity. The Temkin isotherm model is related to the interactions between the adsorbent and adsorbate, and it is calculated by the equation below [23].

$$q_e = BLnC_e + BLnA \qquad (6)$$

$$B = \frac{RT}{b} \qquad (7)$$

where A_T is the binding isotherm at equilibrium conditions, and BT is the heat constant of adsorption. BT shows whether the adsorption process is physical or chemical. The Dubinin-Radushkevich model can be applied to determine and interpret physical and chemical adsorption and can be expressed by [26]:

$$lnq_e = lnq_m - \beta\varepsilon^2 \qquad (8)$$

where q_m the maximum adsorption capacity, and β is the constant for the average adsorption energy. While T is represented by the following equation:

$$\varepsilon = RTln\left(1 + \frac{1}{c_e}\right) \qquad (9)$$

where R is the universal constant for gases (8.314J/mol. K), and T is the temperature. The free energy of transferring 1 mole of the solute to the surface of the adsorbent is referred to as the average energy, E it can be obtained and is represented by the following equation:

$$E = \frac{1}{\sqrt{2\beta}} \qquad (10)$$

The above four models are used to understand the behavior and qualities of the manufactured adsorbent material, because they differ in the following points: Freundlich and Temkin consider heterogeneity, Langmuir assumes a homogenous surface. Langmuir emphasizes monolayer adsorption, Freundlich allows for several layers but does not specify them, Temkin considers interaction effects, and the Dubinin-Radushkevich model is frequently employed for microporous materials. Langmuir and Freundlich make no assumptions about intermolecular interactions, whereas Temkin does. Dubinin-Radushkevich makes assumptions about energy differences between adsorption sites.

5.ADSORPTION KINETICS

Adsorption was studied using kinetic model analysis, which included the pseudo-first-order and pseudo-second-order, Weber-Morris, and Elovich kinetic models. These kinetic models help researchers and engineers understand and predict the adsorption behavior of diverse compounds, which is crucial for applications in environmental remediation, catalysis, and materials science. The pseudo first order is stated linearly in the following equation:

$$\log\ (q_{e-}q_t) = \log\ q_e - \frac{K_1}{2.303}t \qquad (11)$$

where K_1 (1/min) is the pseudo-first-order absorption rate constant, and q_e is the adsorption capacity (mg/g). The pseudo-second-order equation is represented in the following linear form:

$$\frac{t}{q_t} = \frac{1}{h} + \frac{1}{q_e}\ t \qquad (12)$$

As $h = K_2 q_e^2$, where K represents the pseudo-second-order rate constant (g/mg.min).

To understand the adsorption mechanism, the Weber-Morris model (diffusion model within particles) was investigated and determined as follows:

$$q_t = K_{id} t^{0.5} + C \qquad (13)$$

where qt is the adsorption capacity at any given time (mg/g), and K_{id} is the diffusion rate constant within the particles (mg/g.min$^{0.5}$), is the intercept, and t is the time [27]. Elovich model is used to explain the kinetics of chemical adsorption, and can be expressed by the following equation [28]:

$$q_m = \frac{1}{\beta}\ln(\propto \beta) + \frac{1}{\beta} lnt \qquad (14)$$

where (α) is the initial adsorption rate (mg/g), and (β) is the adsorption constant.

The above four models were applied to determine the type of interaction between the adsorbent and the pollutant, they differ in the following points: for rapid surface reactions, the pseudo-first-order model is used, and, for chemisorption scenarios, use the pseudo-second-order model is employed. The Weber-Morris model is excellent for evaluating diffusion effects, and, the Elovich model applies to heterogeneous systems with complex kinetics.

6.ADSORPTION THERMODYNAMICS

The thermodynamic criteria used to assess the feasibility of adsorption include the changes in enthalpy (ΔH°), entropy (ΔS°), and free energy (ΔG°) [29]. They are obtained using the following equations:

$$K_c = \frac{C_{Ae}}{C_a} \qquad (15)$$

$$\Delta G^\circ = {}^{-}RT(K_c) \qquad (16)$$

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \qquad (17)$$

$$\Delta G^\circ = -RTlnKd \qquad (18)$$

Where Kd is the constant of thermodynamics at equilibrium condition, C_{Ae} is the concentration of equilibrium for paracetamol adsorbed on the activated carbon, R is the constant of general gas (8.3143/mol.K), and T is the temperature in Kelvin.

7.RESULTS AND DISCUSSION

7.1.Characterization of Activated Carbon

Figure 3 shows the results of the FTIR examination on the sample of the AC/KOH used in the study. The material has many functional groups. Many peaks were observed. The peak observed at 3394.7 cm^{-1} indicates that it is related to the frequency vibrations of the hydroxyl groups (O-H). The peak at 2918.20 cm^{-1} corresponds to the vibrational frequencies of the methyl groups (C-H), the peak value at 1698.20 cm^{-1} indicates the frequency vibrations (C≡O), which indicates the presence of ketones and carboxylic acids, and the peak at 1538.38 cm^{-1} is attributed to frequency vibrations (N-H), indicating the amides range. The peak at 1417.98 cm^{-1} indicates the presence of alkanes. The peak at 796.89 cm^{-1} is related to the vibrations of aromatic compounds, the peak at 578.64 cm^{-1} may be related to the vibrations of inorganic compounds, and the peaks of the functional groups play a significant role in the binding of adsorbed water [30]. These results are consistent with those calculated by [31]. Figure 4 shows the results of the XRD analysis. The diffraction patterns obtained for activated carbon treated with KOH are presented, it was observed that the material had high density values at 2θ angles of 26.24 degrees, 29.42 degrees, and 29.49 degrees. The high-intensity values indicated the presence of a crystalline carbon structure. It was observed that increasing the values of θ in the spectral pattern, resulted in a decrease in the intensity of the peaks, indicating the presence of an amorphous arrangement of carbon [32]. Figure 5 shows the morphology of activated carbon using a scanning electron microscope (SEM). It shows the changes in the surface structure of the carbon and the development of porosity after activation, carbonization, and treatment with KOH. It is noted that the outer surface contained different and widespread pores. Additionally, it is noted, that the thermal and chemical decomposition contributed to the creation of small pores, which facilitate the adsorption of adsorbed molecules [33].

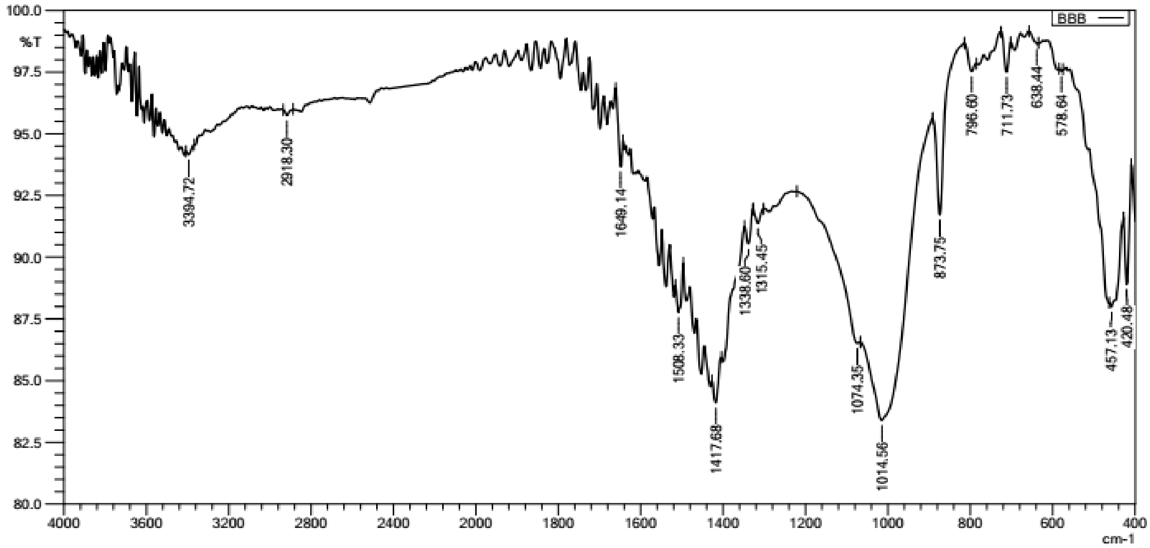


Fig. 3 FTIR Spectra of the Produced Activated Carbon Eichhornia Crassipes Treated with KOH.

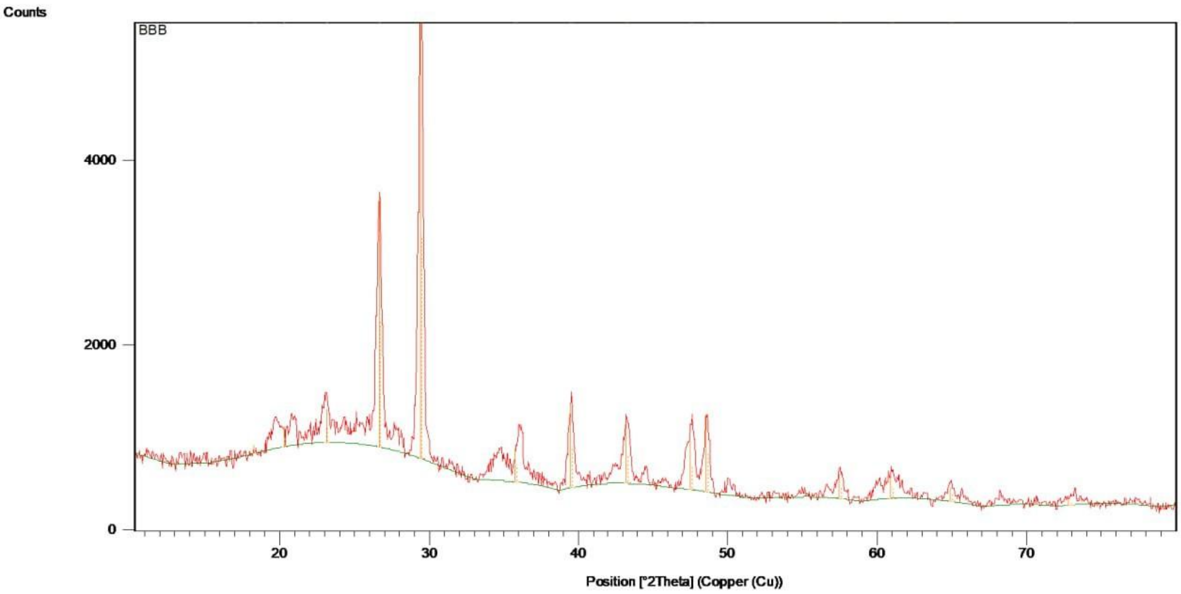


Fig. 4 XRD Analysis Results from Eichhornia Crasspies as AC/KOH Adsorbent.

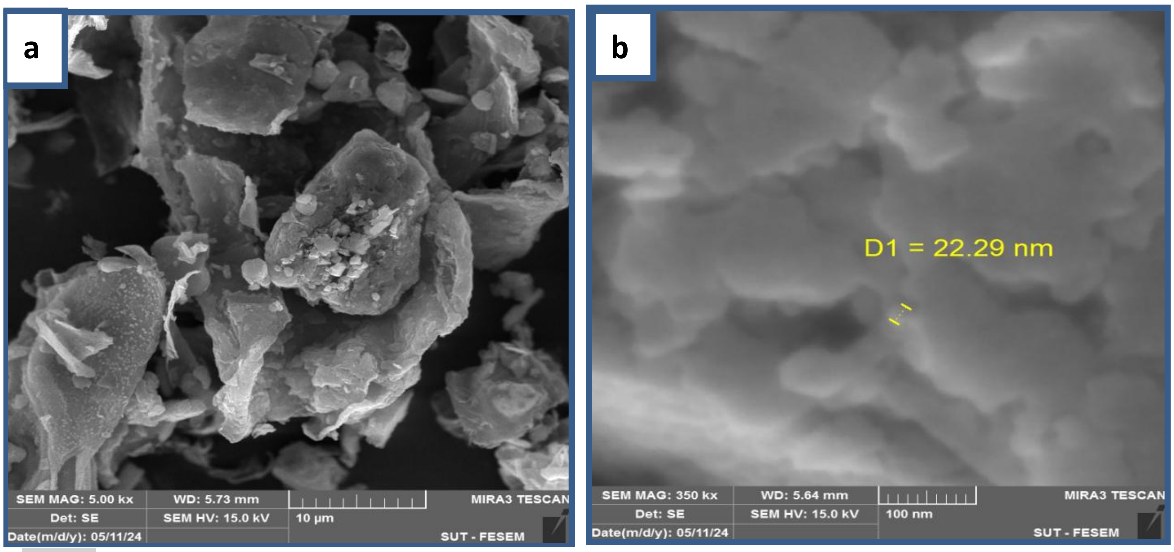


Fig. 5 SEM Image of Activated Carbon of Eichhornia Crasspies at (a) ×5 Magnification, (b) ×350 Magnification.

7.2.Parameters Affecting Adsorption of Paracetamol by AC/KOH

7.2.1.pH Effect

pH is recognized as a key parameter influencing the charge of the solution and the surface of the adsorbent in adsorbent-substance interactions. Effectiveness of the medication, in addition to the different ionic forms presented by the adsorbent species [34]. One of the elements that significantly influenced paracetamol ion absorption was pH. The effect of pH (3-10) was examined using paracetamol concentrations of 10 mg/L, a constant contact time of 180 minutes, and a temperature of 25 °C. As shown in Fig. 6, the experimental findings of AC/KOH demonstrated that high adsorption values were obtained at pH 3. Paracetamol had a clearance efficiency of 74% at 10 mg/L, 70% at 30 mg/L, and 53% at 50 mg/L, as shown in Fig. 7. The concentration of anions on the surface of the adsorbent grows as pH affects the adsorption process by dissociating functional groups on active sites on the adsorbent surface, resulting in a large number of negative sites. The effectiveness of paracetamol elimination declines at a pH of (4-10). Increasing the pH results in a hydrodynamic repulsion between negatively charged adsorbents and paracetamol anions, affecting removal efficiency. These results are consistent with those reported by Abdulrahim [16] and [35].

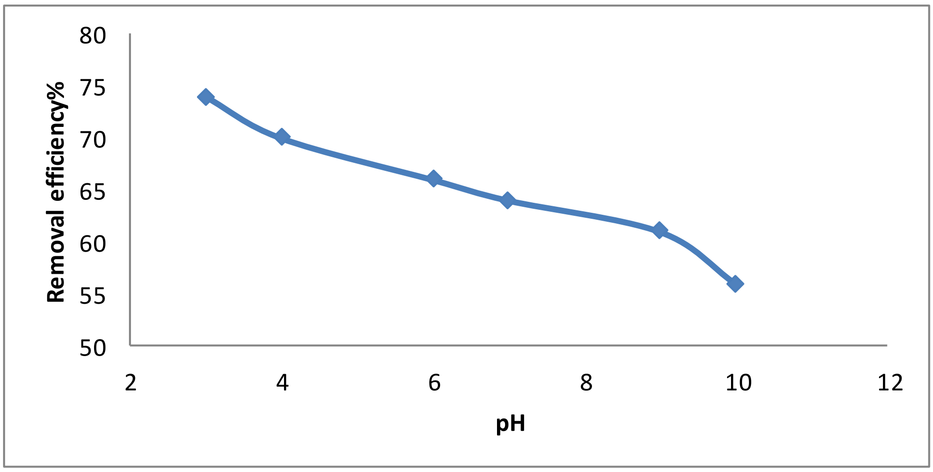


Fig. 6 Effect of pH on Paracetamol Removal Efficiency Into AC/KOH (Paracetamol Initially Concentration of 10 mg/L, Temp. of 20 °C, and AC/KOH Dosage of 2 g/L, Agitation Speed 150 rpm).

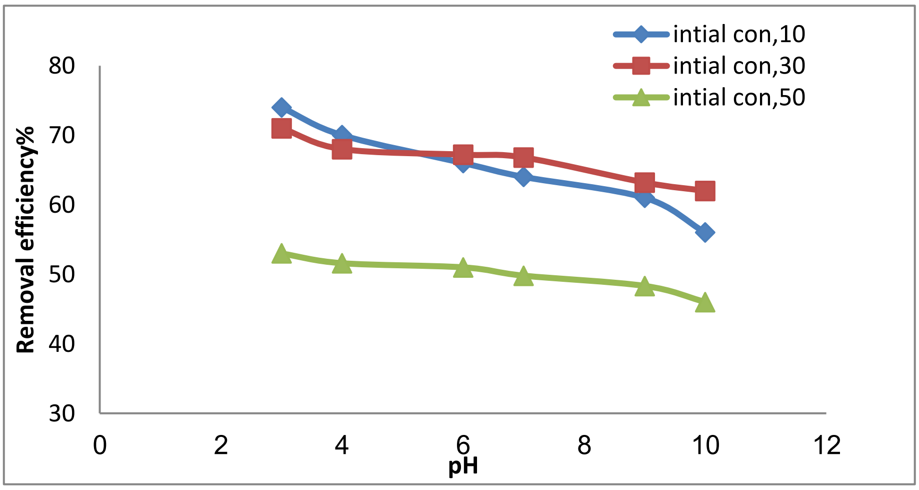


Fig. 7 Effect of pH on the Removal Efficiency of Paracetamol at Initial Concentrations (10,30,50 mg/L, AC Treated with KOH and Dose = 1 g/L, and Contact Time = 180 min).

7.2.2.Adsorbent Dosage Effect

The adsorbent dose is a significant component in evaluating the adsorption process, and providing the adsorbent necessitates appropriate testing since the provision of active adsorption sites is a critical aspect that significantly impacts removal effectiveness [36]. The influence of the AC/KOH dosage on the absorption phenomena was investigated using different doses of AC/KOH (0.2-2.5 g/L) to an adsorbent concentration of 10 mg/L, while keeping all other factors constant. The results, on the other hand, revealed an inverse relationship between adsorption efficiency and adsorption capacity, as shown in Fig. 9. Adsorption effectiveness increased with increasing absorbent dose, reaching 78.38% at an optimum dose of 2 g/L; however, adsorption capacity declined from 30.34 mg/g to 1.683 mg/g, as shown in Fig. 8. The efficacy of pollutant removal increased with the adsorbent due to the increased number of active sites available on the material's surface. This result means that there are more opportunities to bind pollutants or target molecules, increasing the likelihood of interaction between the pollutant and the adsorbent. The more adsorbent there is, the more likely it is that pollutant molecules will collide with the active sites, resulting in a drop in pollutant concentration in the solution. This drop in concentration improves the dynamic processes of delivering pollutants to the surface, and it increases the availability of larger surface areas for the adsorbent to accommodate more pollutant molecules, thereby increasing the effectiveness of removal [15, 37].

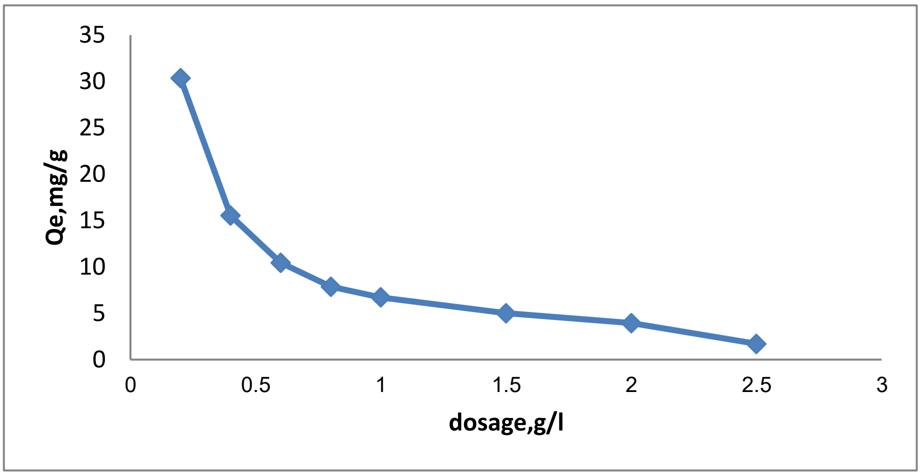


Fig. 8 Effect of AC/KOH Dose on the Absorption Capacity of Paracetamol (Paracetamol Initial Concentration of 10 mg/L, Temp. of 25 °C, pH=3, Contact Time=180 min, and Agitation Speed 200 rpm).

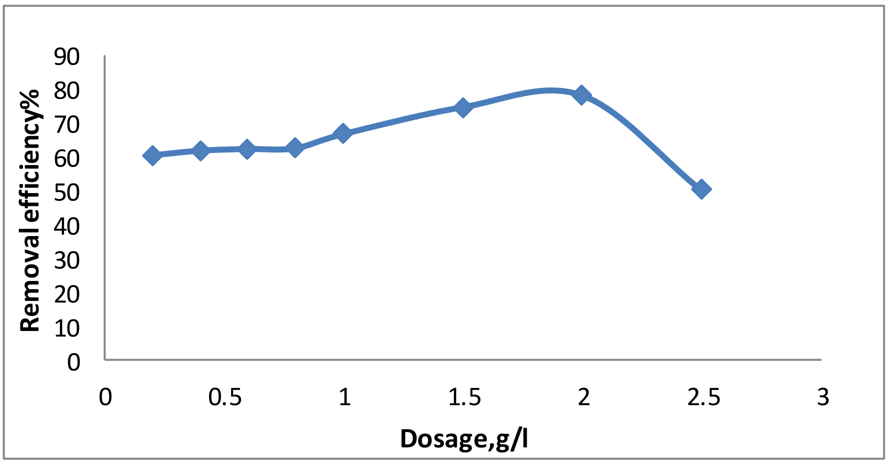


Fig. 9 Effect of AC/KOH Dose on Paracetamol Removal Efficiency (Paracetamol Initial Concentration of 10 mg/L, Temp. of 25°C, pH=3, Contact Time=180 min, and Agitation Speed 200 rpm).

7.2.3.Contact Time Effect

Contact time is a crucial criterion for evaluating the applicability of the adsorption process. It can either positively or negatively affect the dynamics of adsorption and removal of medicinal ions. Figure 10 demonstrates the impact of contact time on ion adsorption capability. The Pharmacokinetics of paracetamol were studied initial concentrations of 10 mg/L for AC/KOH and a contact time of 15-240 minutes, with all other parameters maintained constant. Calculations show that the removal rates, R%, are equivalent to the adsorption capacity, q_e. The results showed that the rate of adsorption was relatively low during the first two hours of the process, however, it increased after 120 minutes, reaching 70.04% at a concentration of 10 mg/L. After 180 minutes, the equilibrium stage was reached, resulting in the optimum concentration for removal efficiency of 10 mg/L (75%). Following that, there was no additional rise in absorption, and equilibrium was reached at 180 minutes. This behavior is attributed the initial increase in removal rate resulting from the availability of more active surface sites, which decreased as adsorption progressed [15], rather than to the porous nature of activated carbon. However, from 180 to 240 minutes, the adsorption rate decreased, and saturation occurred with the passage of time, indicating that there was no further increase in the absorption field [38]. It is not possible to introduce more adsorbed molecules of paracetamol to the inner surface of the carbon due to the high degree of saturation ,and the molecules occupy all the free spaces [39].

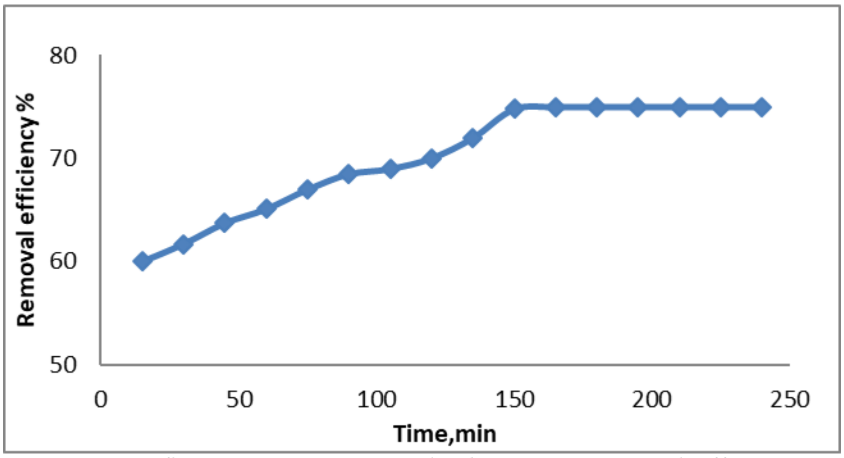


Fig. 10 Contact Time Influence on Paracetamol Adsorption Removal Efficiency in (the Initial Concentrations Paracetamol 10 mg/L, pH= 3, Dose AC/ KOH = 2 g/l. Agitation speed 150 rpm, and Temp 25 ℃).

7.2.4.Initial Concentration Effect

The impact of the initial paracetamol concentrations on adsorption capacity is very important because equilibrium adsorption capacity Qe increases directly with the initial concentrations of the pharmaceutical substances. Figure 11 shows the relationship between the initial concentrations of paracetamol (10,30,40,60, and 80 mg/L) for AC/KOH). Adsorption capacity increased with concentration. The process continued until all accessible active sites on the activated carbon were filled [40]. In terms of elimination, pharmaceutical molecules were more firmly connected to the material. Adsorption in active sites occurred due to the presence of attractive forces (static electricity, and Van der Waals forces), and the adsorption efficiency increased. At a dosage of 10 mg/L, the highest removal rate was 75.91%, while at 80 mg/L, the lowest removal rate was 36.83%. This result indicates that as concentrations increased, the elimination rate decrease resulting from a reduction in attractive forces and a decrease in the availability of free active sites [41].

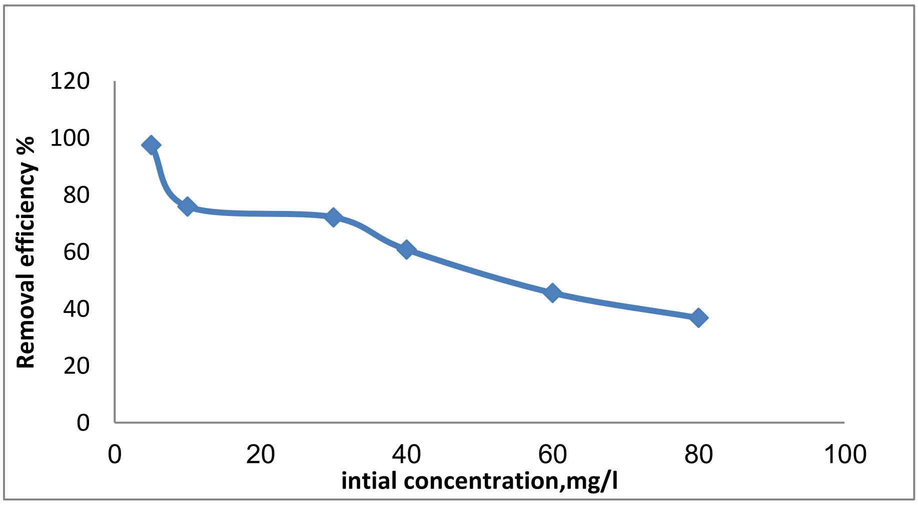


Fig. 11 The Effect of Initial Paracetamol Concentration on Removal Efficiency. (AC/KOH Doses of 2 g/l, pH = 3, Contact Time = 180min. Agitation Speed 200 rpm, and Temp 25 ℃).

7.2.5.Shaking Speed Effect

The speed of shaking affects the adsorption process. According to studies, each absorbent substance has an optimal speed that must be validated. Figure 12 depicts the effect of the number of revolutions per minute on the absorption of medicinal materials (paracetamol) at various stirring speeds (100, 150, and 200 rpm) for AC/KOH produced from Eichhornia Crassipes. The results demonstrated that the removal rates increased with speed [42] until they approached equilibrium. The highest removal efficiency was 72.76% at a rotation speed of 200 rpm. This result occurs due to increased turbulence around the particles, which is considered in the degree of thickness of the boundary layer around the particle's adsorbent. [43]. It should be noted that a further increase in shaking speed intensifies turbulence around the particles and produce reverse vortices, which negatively affect the adsorption process [44].

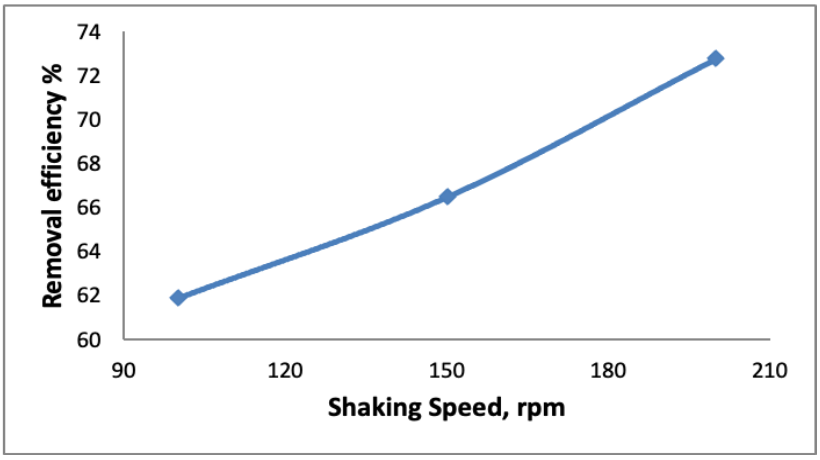


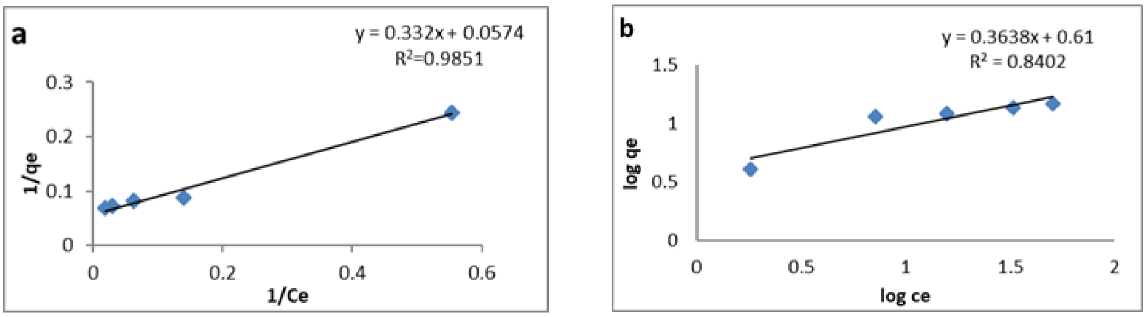
Fig. 12 The Effect of Shaking Speed on Removal Efficiency (AC/KOH Doses of 2g/l, pH = 3, Contact Time of 180 min, Initial Concentration of Paracetamol 10 mg/L, and Temp 25 C°).

7.3.Adsorption Models

It is a strategy for selecting the optimal model for linear regression analysis, while also explaining the distribution of pollutants on the absorbent material and the related adsorption mechanism [23]. These isotherm models study the interactions between the adsorbent and the adsorbate utilizing equilibrium analysis of the adsorption process [45]. Additionally, the interactions between paracetamol and AC/KOH prepared from the Eichhornia Crassipes plant are described. Isotherm studies were conducted using (Langmuir, Freundlich, Temkin, and Dubinin-Radushkevich) models. Parameters and data obtained, along with an assessment of the suitability of the isotherms, were used to accurately examine and determine the adsorption [25]. The Langmuir and Freundlich models were applied, as shown in Fig. 13 and Table 1, to analyze the equilibrium data and explain the adsorption process. All values and parameters were included in the regression analysis, indicating that the data align best with the specific studies. The highest value of R^2 was calculated at 0.985 in the Langmuir isotherm indicating that the results essentially follow the Langmuir model. The adsorption was very suitable between paracetamol and activated carbon treated with KOH. Therefore, the Langmuir model is more suited for physical adsorption. When comparing these models in this study, it was found that they follow the Langmuir model, the adsorption was single-layer, and the isotherms were arranged according to decreasing R^2, which is Langmuir isotherms > Temkin isotherms > Freundlich isotherms > Dubinin-Radushkevitch , as presented in [27, 17].

Table 1 Details of Paracetamol Adsorption by AC/KOH Using Isotherm Models.

	Adsorption Models	Parameters	Value	R^2
AC treated with KOH	Langmuir	qm (mg/g)	17.42160279	0.9851
		K_c (L/mg)	0.172891566	
		RL	0.366445916	
	Freundlich	1/n	0.3638	0.8402
		K_f (mg/g)	4.073803	
	Temkin	B_1	3.0338	0.915
		LnK_t	3.171995	
	Dubinin-Radushkevitch(D-R)	qm (mg/g)	13.3244	0.8168
		K (mol^2/J^2)	0.000001	
		E (J/mol)	707.1068	



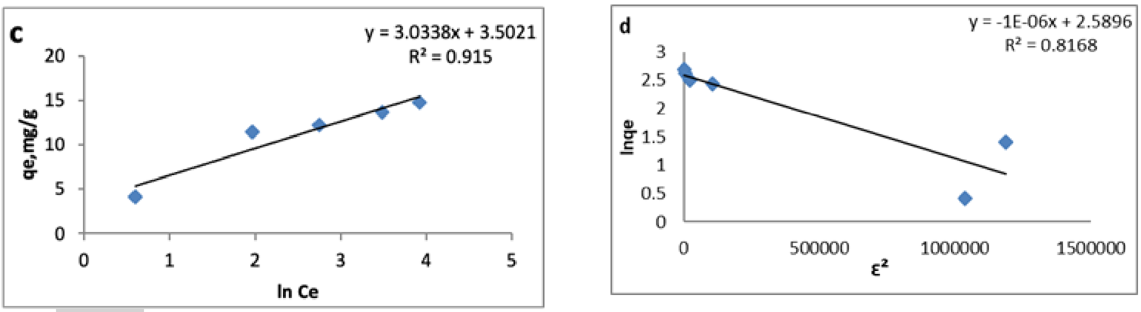


Fig. 13 (a) Langmuir Model, (b) Freundlich Model, (c) Temkin Model, and (d) Dubinin-Radushkevich Model for Paracetamol Solution:(Initial concentration = 10 mg/L, pH = 3, AC/KOH dose = 2 g/l, Time = 180 min, Solution volume = 50 mL. Agitation speed 200 rpm, and Temp 25 C°).

7.4.Adsorption Kinetics

Using various adsorption kinetic models enables researchers to gain a comprehensive understanding of the adsorption process under diverse conditions. Each model incorporates distinct assumptions and equations, making it applicable to a variety of systems and providing insights into various adsorption mechanisms. The kinetic study measures the rate of adsorption, which is calculated using models of adsorption kinetics, such as the pseudo-first-order, pseudo-second-order, Weber-Morris, and Elovich models. Linear curves based on model equations and calculated coefficients, as shown in Fig. 14 and Table 2, where the method of absorption is described. AC/KOH from the Eichhronia Crassipes plant indicated the linear kinetic model values. According to the results, the pseudo-second-order kinetic model was the best and most appropriate for the experimental data, yielding an R^2 value= of (0.998). It was the closest to the experimental value (0.99) compared to the other models, demonstrating that the adsorption process fits the pseudo-second-order kinetics model [16, 15]. The pseudo-second-order model is very good for revealing strong interactions and providing insights into the underlying adsorption mechanisms, making it a valuable tool in various scientific and industrial applications.

Table 2 Adsorption Kinetics Details of Paracetamol Adsorption by AC/KOH) at Initial Concentration = 10 mg/L.

Type of AC	Parameters	Pseudo- first-order	Pseudo-second-order	Weber–Morris	Elovich
AC treated with KOH	R^2	0.7864	0.9987	0.9155	0.929
	Constant,	K_1= -0.0001594	K_2=6.74502466	K_{di}=0.0031	α=2067.61 β=4.008
	Qe	0.27599423	3.70096225	____	___

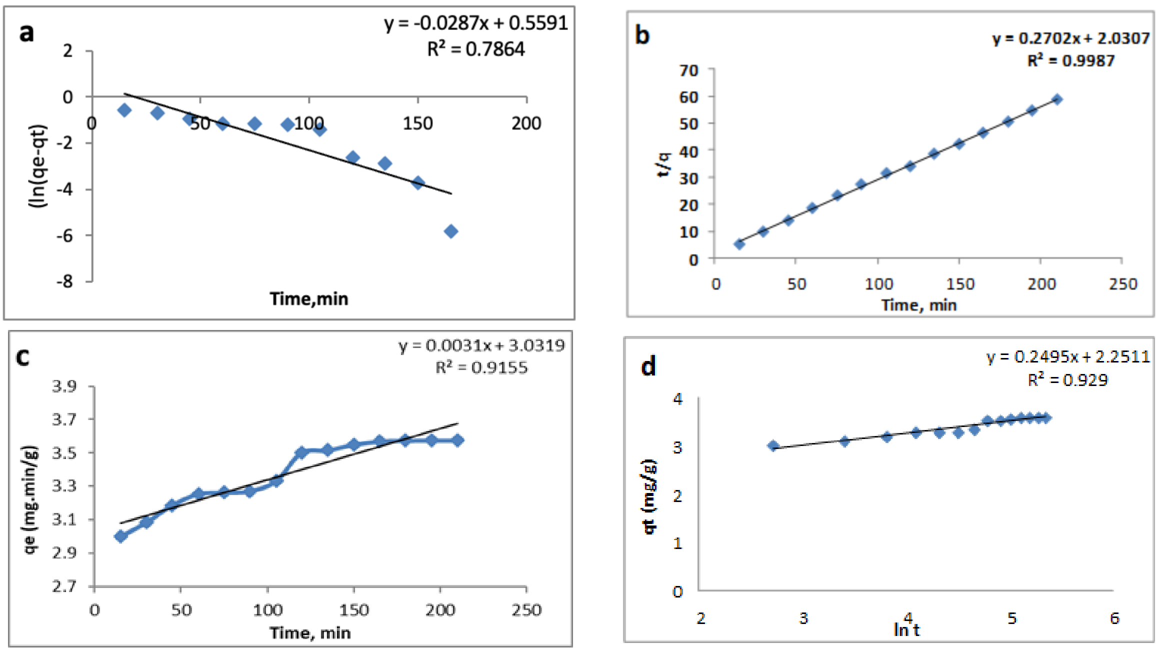


Fig. 14 (a) Pseudo-First-Order, (b) Pseudo-Second-Order, (c) Weber-Morris, and (d) Elovich Models to Examine the Adsorption Behavior of AC/ KOH for Paracetamol Solution.

7.5.Thermodynamic Parameters

The thermodynamic parameters ΔG, ΔH, and ΔS, measured using Eqs. (16, 17, and 18), can determine the direction and feasibility of chemical and physical adsorption reactions. Table 3 shows that negative ΔG values varied with temperature (-20 to 40 kJ/mol) and adsorbent material (AC/KOH). Negative ΔG values indicate that the adsorption reaction can occur spontaneously [47]. The negative enthalpy values (ΔH) show physical and exothermic adsorption between paracetamol and AC/KOH. Furthermore, the negative entropy value (ΔS) suggests enhanced bonding between molecules and materials. The pharmacological qualities of AC/KOH, as well as the ease with which bonds can be formed, suggest that the entropic effect drives the adsorption operation [16].

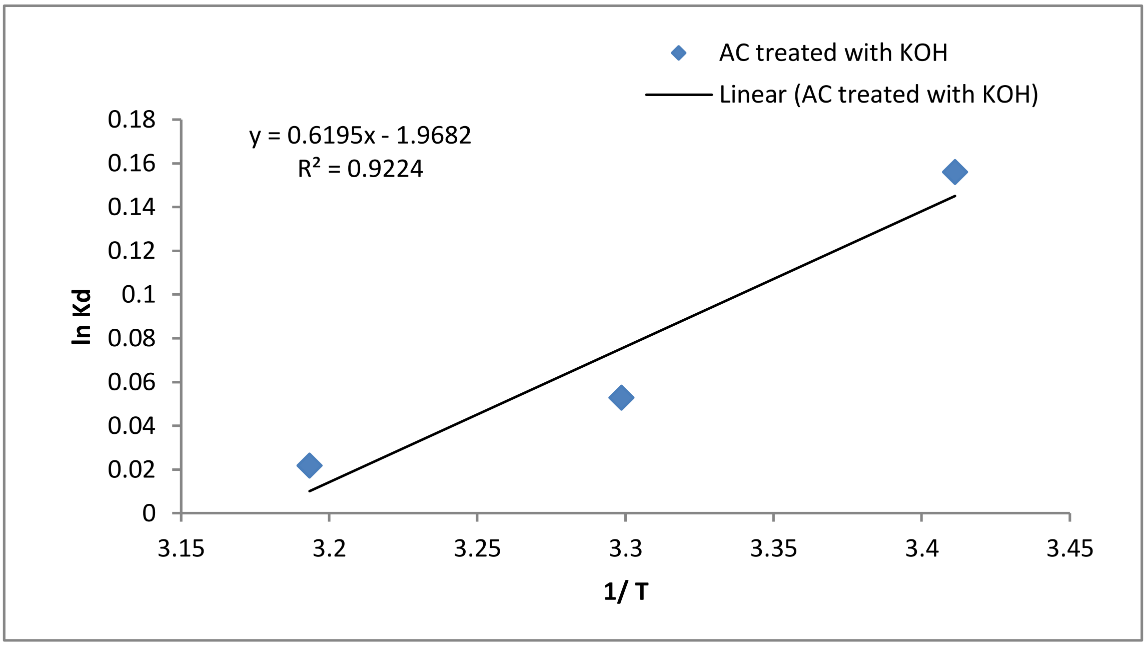


Fig. 15 The Plot of ln KL vs. 1/T for Paracetamol Sorption Onto (AC Treated with KOH).

Table 3 Thermodynamic Parameters for Paracetamol Adsorption onto AC/KOH.

	Temp. (k)	K_d	ΔG°(kJ/mol)	ΔH°(kJ/mol)	ΔS°(J./mol.K)	R^2
AC/KOH	293.15	1.168	-0.38035	-5.15052	-4.4464	0.9224
	305.15	1.054	-0.13313			
	313.15	1.0220	-0.05684			

8.CONCLUSION

Since pharmaceuticals are widely used and have a long-lasting active mechanism in aquatic systems, they are considered potentially harmful pollutants that pose a significant risk to both humans and aquatic life. Therefore, pharmaceutical-containing wastewater should be treated before being released into the environment. Under ideal conditions, it was demonstrated that activated carbon made from Eichhronia Crassipes and treated with KOH could remove paracetamol from aqueous solutions with an efficiency of 81.91%. The Langmuir model provided a better fit for the adsorption data, as evidenced by the correlation coefficient (R^2 = 0.9851), which suggests that the isothermal model is applicable. The pseudo-second model provides the best description of kinetic adsorption. According to the present results, the prepared adsorbent is a promising, inexpensive, eco-friendly, and efficient material for removing paracetamol. It can also be used to assess the efficacy of the material by removing other kinds of pharmaceutical waste.

ABBREVIATIONS

(BET)	(Brunauer-Emmett-Teller)
FTIR	Fourier-transform infrared spectroscopy
XRD	X-Ray photoelectron spectroscopy
SEM	Scanning Electron Microscope
Greek symbols	
(α)	represents the initial adsorption rate
(β)	adsorption constant

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