

Eco-adsorbents at work: Mechanistic and isotherm analysis of oxalic acid adsorption on coconut shell and bamboo charcoal

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Abstract- This study focuses on the adsorption of oxalic acid from an aqueous solution onto coconut shell and bamboo charcoal, which are both low-cost and eco-friendly adsorbents. The Langmuir isotherm model gave good linearity, hence demonstrating monolayer adsorption on homogeneous surface sites. The correlation coefficients were R^2 is 0.9472 for coconut shell and 0.9764 for bamboo charcoal, confirming the suitability of the Langmuir model. By calculating the Langmuir constants (K_L), it was found that bamboo charcoal has a higher adsorption affinity, while coconut shell charcoal has a higher monolayer adsorption capacity. The thermodynamic analysis has presented negative standard gibbs free energies (ΔG_{ads}^0) of $-15.48 \text{ kJ mol}^{-1}$ and $-19.34 \text{ kJ mol}^{-1}$ for coconut shell and bamboo charcoal, respectively, hence proving the spontaneity and feasibility of the adsorption process, which was governed by physisorption. The more negative ΔG_{ads}^0 , and higher K_L values, the stronger the adsorbate-adsorbent interactions and the superior the performance. These findings support the potential of these biochar-based materials as sustainable adsorbents towards the removal of oxalate pollutants from industrial effluents and offer an environmentally friendly approach for wastewater management.

Keywords: oxalic acid, coconut shell charcoal, bamboo charcoal, adsorption equilibrium constant, Gibbs free energy.

I. INTRODUCTION

Adsorption is a modern surface and environmental chemistry phenomenon of prime importance, finding wide applications in wastewater treatment, environmental remediation, catalysis, separation science, and various analytical technologies. It is commonly defined as the adhesion of atoms, ions, or molecules (adsorbate) from a fluid phase onto the surface of a solid or liquid (adsorbent), usually forming a molecular layer. Unlike absorption, in which the substance diffuses into the bulk of the material, adsorption is strictly a surface-governed interaction, usually controlled by Van der Waals forces, electrostatic interactions, hydrogen bonding, or chemisorption. Indeed, this surface specificity now makes adsorption indispensable for processes in which the interactions between molecules at interfaces determine the efficiency of separation, purification, or catalytic activity [1].

Among all classes of adsorbents, carbon-based materials, specifically activated charcoal, have attracted great interest [2]. Activated charcoal possesses a highly porous framework, high surface area, and tunable pore size distribution in terms of micro-, meso-, and macropores with diverse surface functionalities, making it highly competent for the removal of organic acids, dyes, toxic gases, heavy metals, and trace contaminants from water. In addition, low cost, natural abundance, chemical inertness, ease of regeneration, and environmental compatibility add to its usability on an industrial scale [3].

Oxalic acid, a small dicarboxylic acid represented as $\text{C}_2\text{H}_2\text{O}_4$, is universally distributed in nature and finds extensive applications in industries [4]. It is found in spinach, beets, cocoa, tea leaves, and rhubarb, and industrially, in textile processing, metal polishing, rust removal, leather tanning, and as a reducing agent. Although useful, it becomes an environmental hazard upon discharge into the water systems through untreated effluents from textile mills, paper and pulp industries, dyeing units, agricultural runoff, and electronic waste recycling processes [5].

The high acidity and water solubility of oxalic acid allow it to readily dissociate and act as an agent for acidification in soils and aquatic bodies. This forms highly insoluble calcium oxalate upon reaction with calcium ions, which causes kidney stones in humans, metabolic disturbances, nutritional deficiencies in plants due to nutrient deficiencies, and toxicity in livestock [6]. Besides this, oxalic acid increases the mobility of heavy metals such as lead, cadmium, and chromium in both soil and water, leading to increased ecological risks [7].

Remediation strategies explored include catalytic wet air oxidation (CWAO), catalytic ozonation, biological degradation, membrane separation, ion exchange, and adsorption [8]. Most of them achieved a high degradation efficiency, except that CWAO and ozonation normally consume high energy due to the complex operation and expensive catalysts. The biological methods are environmental friendly but also face challenges such as slow degradation rates and sensitivity toward pH, temperature, and toxic by-products [9]. In contrast, adsorption has been considered an efficient and sustainable treatment due to its ease of operation, a

high removal efficiency at low pollutant concentrations, a small amount of sludge generation, and a potential for the regeneration of adsorbents [10].

In recent years, low-cost, bio-derived adsorbents, including coconut shell charcoal and bamboo charcoal, have been considered as alternatives to commercially available activated carbon [11-14]. Due to the presence of a well-developed microporous structure, high carbon content, superior mechanical strength, and excellent thermal stability, coconut shell charcoal is highly effective in the adsorption of organic pollutants of low molecular weight, such as oxalic acid [15]. Bamboo charcoal, resulting from fast-growing biomass such as bamboo, possesses a mixture of mesoporous and macroporous networks with numerous hydroxyl and carboxyl surface functional groups, is lightweight, biodegradable, and environmentally sustainable [16].

Ikram et al. investigated a cost-effective biosorbent in the form of chicken eggshells, used for the removal of inorganic and organic impurities from aqueous solutions. The thermodynamic results indicated that the processes of adsorption were spontaneous and favorable. Equilibrium data were subjected to the Freundlich, Langmuir, Temkin, and Harkins-Jura isotherms to better represent the nature of adsorption [17]. Similarly, H. Foya investigated the adsorption of oxalic and maleic acids onto activated carbons derived from tamarind seeds. The adsorption process was found to be predominantly governed by chemisorption, occurring on both homogeneous and heterogeneous surface sites, accompanied by intraparticle diffusion [18]. Lee et al. investigated the adsorption of oxalic, malonic, and succinic acids onto activated carbon at 5 °C and 40 °C. Langmuir and the Freundlich models were applied for the analysis of the equilibrium data. According to the data obtained by them, the potential theory fitted best, giving the least deviation (1.80–5.44%), and they successfully developed a generalized adsorption curve for these dicarboxylic acids [19]. Although numerous studies have investigated the adsorption of organic acids onto activated carbon, comparative analyses of oxalic acid adsorption on coconut shell-derived charcoal and bamboo-derived charcoal under controlled conditions remain limited. Therefore, the present study aims to provide a comprehensive evaluation of their adsorption performance, including efficiency, equilibrium behavior, kinetic characteristics, and mechanistic aspects.

This work seeks to address the existing research gap through a systematic investigation of oxalic acid adsorption onto coconut shell and bamboo charcoals using various adsorption isotherm models, namely Langmuir, Freundlich, Temkin, El-Awady, Frumkin, and Flory–Huggins. Emphasis is placed on understanding the adsorption mechanism, equilibrium modeling, kinetic and thermodynamic parameters, as well as the influence of varying initial oxalic acid concentrations. The experimental findings offer a comparative assessment of the adsorption capacities of both adsorbents, highlighting their potential applicability as low-cost, sustainable, and environmentally friendly materials for the treatment of oxalic acid-contaminated industrial and e-waste effluents [20].

II. Materials and experimental procedure

The current study was conducted to investigate the adsorption characteristics of oxalic acid onto various charcoal-based adsorbents in a controlled laboratory setting. Throughout the experimentation, analytical-grade oxalic acid dihydrate ($C_2H_2O_4 \cdot 2H_2O$), sodium hydroxide (NaOH), coconut shell charcoal powder, bamboo charcoal powder, and distilled water were used. Both the charcoal samples were procured locally and finely powdered for homogeneity and maximum surface exposition [21].

A 0.1 N NaOH solution was prepared and standardized. Oxalic acid solutions (2%, 4%, 6%, 8% and 10% w/w) were prepared by dissolving the required amounts of oxalic acid dihydrate in distilled water and diluting to 100 mL in volumetric flasks. Aliquots of 40 mL from each solution were transferred into 100 mL beakers for subsequent adsorption experiments. To each beaker, 4 g of the respective adsorbent (coconut shell charcoal or bamboo charcoal) and a magnetic stir bar were added. The suspensions were stirred at 300 rpm and 27 ± 1 °C for 3 hrs to ensure sufficient contact between the oxalic acid molecules and the adsorbent surface. After equilibration, the mixtures were filtered through standard filter paper to separate the residual charcoal particles [22].

The residual concentration of oxalic acid in the filtrate was determined by acid–base titration. A 5 mL aliquot of each filtrate was titrated against standardized 0.1 N NaOH after the addition of 1–2 drops of phenolphthalein indicator as the endpoint marker. The amount of oxalic acid adsorbed at each concentration was calculated from the difference between the initial and residual concentrations, enabling the determination of adsorption efficiency and adsorption capacity [23]. This procedure ensured reliable correlation between oxalic acid concentration and adsorption performance, allowing a comparative evaluation of coconut shell and bamboo-derived charcoals.

III. Results and discussion

In the present study, a systematic evaluation of oxalic acid adsorption onto coconut shell and bamboo charcoal was carried out using batch experiments. Fittings for several isotherm models namely, Langmuir, Freundlich, Temkin, El-Awady, Frumkin, and Flory–Huggins were made. The application of a variety of models to experimental data enabled the identification of the constants of isotherms and standard Gibbs free energy (ΔG_{ads}^0), which gives information on heterogeneity of the surface, adsorption capacity, and spontaneity of the process. Results obtained showed completely different behaviors with respect to both adsorbents depending on the concentration of oxalic acid, the nature of their surfaces, and temperature [24].

Comparison among thermodynamic and kinetic parameters provided more information with regard to the mechanisms that rule the adsorption process, once more evidencing the viability and efficiency of low-cost biochar materials tested for oxalic acid removal [25]. The results obtained from the experimental procedure are presented in Tables 1 and 2 for coconut shell charcoal and bamboo charcoal, respectively. Here, C represents the initial concentration of oxalic acid expressed in percent, V denotes the volume of standardized 0.1 N NaOH required to titrate 5 mL of the oxalic acid solution after adsorption, C_i is the initial concentration of oxalic

acid in mol L^{-1} , C_e is the equilibrium concentration of oxalic acid in mol L^{-1} , x corresponds to the amount of oxalic acid adsorbed, and m is the mass of charcoal. Adsorption capacity is represented by Q_e , which is the ration of x to m . Using equation (1), the value of x was calculated [26]:

$$x = \frac{(C_i - C_e) \times 40}{1000} = \frac{(C_i - C_e)}{25} \quad (1)$$

Table 1: Adsorption data showing C_i , C_e , x , and Q_e of oxalic acid on coconut shell charcoal at varying adsorbent doses.

Experiment No.	C (%)	V (mL)	C_i (mol L^{-1})	C_e (mol L^{-1})	x (mol)	Q_e (mol g^{-1})
1	2	5.2	0.3174	0.0799	0.0095	0.002375
2	4	18.2	0.6349	0.2799	0.0142	0.00355
3	6	39.0	0.9523	0.5998	0.0141	0.003525
4	8	49.0	1.2698	0.7536	0.0207	0.005162
5	10	83.4	1.5873	1.1426	0.0178	0.004447

Table 2: Adsorption data showing C_i , C_e , x , and Q_e of oxalic acid on bamboo charcoal at varying adsorbent doses.

Experiment No.	C (%)	V (mL)	C_i (mol L^{-1})	C_e (mol L^{-1})	x (mol)	Q_e (mol g^{-1})
1	2	5.0	0.3174	0.0769	0.0096	0.002405
2	4	9.0	0.6349	0.1384	0.0199	0.004965
3	6	52.2	0.9523	0.7151	0.0095	0.002372
4	8	73.3	1.2698	1.0042	0.0106	0.002656
5	10	81.0	1.5873	1.2450	0.0137	0.003423

Langmuir Isotherm

The Langmuir isotherm explains monolayer adsorption on a homogeneous surface with a finite number of identical sites, assuming uniform energy and no interaction between adsorbed molecules. It is expressed as equation (2):

$$\frac{C_e}{Q_e} = \frac{1}{K_L Q_{max}} + \frac{C_e}{Q_{max}} \quad (2)$$

where Q_{max} represents maximum adsorption capacity, and K_L indicates adsorption affinity or equilibrium adsorption constant [27]. Its advantages include simplicity and suitability for describing chemisorption, while its limitations arise from ignoring surface heterogeneity, multilayer formation, and adsorbate interactions.

$\frac{C_e}{Q_e}$ vs C_e gives a straight line where the slope gives the value of $\frac{1}{Q_{max}}$ and the intercept gives $\frac{1}{K_L Q_{max}}$ from which K_L value can be calculated [28].

Freundlich Isotherm

The Freundlich isotherm is an empirical model that explains adsorption on heterogeneous surfaces with sites of varying affinities. It assumes that adsorption occurs first on high-energy sites and progressively on lower-energy ones as surface coverage increases. The model is expressed by the equation (3) and its logarithm form by equation (4):

$$Q_e = K_F C_e^{1/n} \quad (3)$$

$$\log Q_e = \log K_F + \frac{1}{n} \log C_e \quad (4)$$

where K_F represents adsorption capacity, and $\frac{1}{n}$ reflects adsorption intensity [29]. Thus, according to Eq (4), a graph of $\log Q_e$ vs $\log C_e$ gives the value of n and K_F . The Freundlich isotherm effectively describes multilayer adsorption on non-uniform surfaces, but it lacks a theoretical saturation limit and is less accurate at high concentrations [30].

Temkin Isotherm

The Temkin isotherm accounts for adsorbate–adsorbent interactions by assuming that the heat of adsorption decreases linearly with increasing surface coverage due to these interactions and it is represented by the equation (5):

$$Q_e = \frac{RT}{b_T} \ln A + \frac{RT}{b_T} \ln C_e \quad (5)$$

where R is the gas constant, T is the temperature, b_T is the Temkin isotherm constant and is dimensionless. $\frac{RT}{b_T}$ is a constant related to heat of adsorption, and A is the equilibrium binding constant [31]. Q_e vs $\ln C_e$ gives the trend line whose slope and intercept give the values of A and b_T parameters. The Temkin model can also explain the influence of the adsorbate-adsorbent interactions, but this model is less useful for strongly heterogeneous systems and multilayer adsorption [32].

Flory Huggins Isotherm

The Flory-Huggins isotherm accounts for adsorption by incorporating both a factor for degree of surface coverage and one for the role of the solvent. It is uniquely well suited to applications where the adsorbate molecule size and the displacement of solvent molecules both play an important role in adsorption. This model can be expressed as equation (6):

$$\log \frac{Q_e}{C_e} = \log K + x \log(1 - Q_e) \quad (6)$$

where Q_e is the surface coverage, K is the Flory-Huggins equilibrium constant, and x is the size parameter, giving the number of surface sites occupied by one adsorbed molecule [33]. Thus, a plot between $\log \frac{Q_e}{C_e}$ vs $\log(1 - \theta)$ gives a straight line whose slope gives the value of x and intercept K. This isotherm gives information on the adsorption process concerning spontaneity but has serious limitations in the case of non-ideality of the system [34].

Frumkin Isotherm

The Frumkin isotherm describes adsorption by introducing lateral interactions between the adsorbed molecules. This makes it more realistic compared with the Langmuir model for systems where intermolecular forces are relevant. It is expressed as equation (7):

$$\frac{Q_e}{1-Q_e} = KC_e \exp 2aQ_e \quad (7)$$

Take the natural log and rearrange the equation (7):

$$\ln \frac{Q_e}{(1-Q_e)C_e} = \ln k + 2aQ_e \quad (8)$$

where k is the adsorption constant, and a is the interaction parameter, which denotes the nature of adsorbate interactions-positive for attraction and negative for repulsion [35]. This model effectively accounts for non-ideal adsorption behavior but is mathematically complex and less suitable for highly heterogeneous surfaces [36].

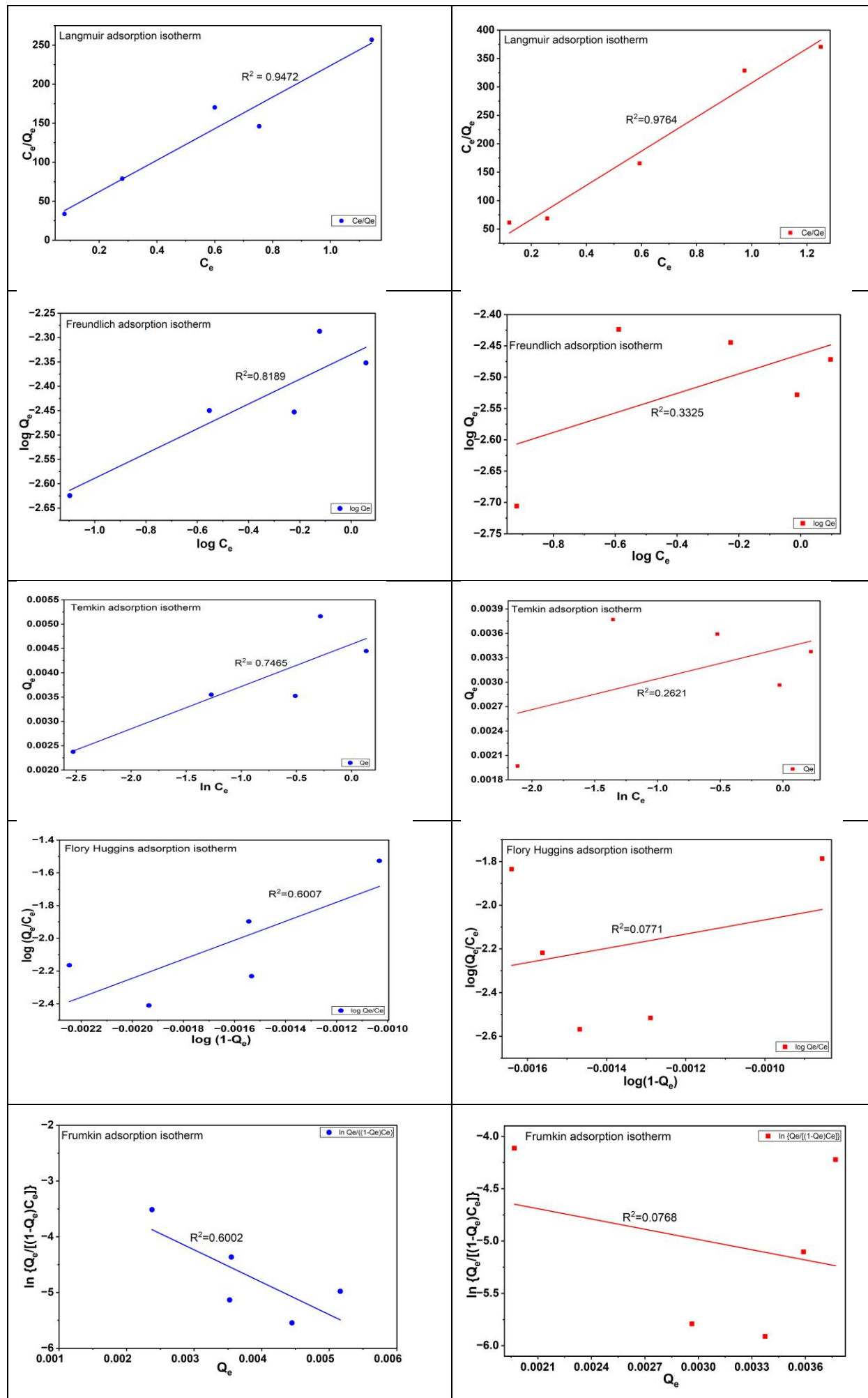
El-Awady Isotherm

The El-Awady isotherm model has been widely applied to study the adsorption process involving complex mechanisms, especially in corrosion inhibition systems. This isotherm relates the fraction of surface coverage to the binding constant and the number of adsorbate molecules occupying each active site. The model is expressed as equation (9):

$$\log \frac{Q_e}{1-Q_e} = \log K + y \log C_e \quad (9)$$

y is the number of adsorbate molecules per active site, and K is the equilibrium constant [37]. Plot of $\log \frac{Q_e}{1-Q_e}$ vs $\log C_e$ gives a straight line, from which the value of both the constants can be evaluated. This model provides valuable insight into the nature of adsorption, that is, multilayer or monolayer, and the strength of adsorbate-adsorbent interactions [38]. Different adsorption isotherms were analyzed and tabulated in Table 3.

Adsorption Isotherms for coconut shell charcoal	Adsorption Isotherms for bamboo charcoal
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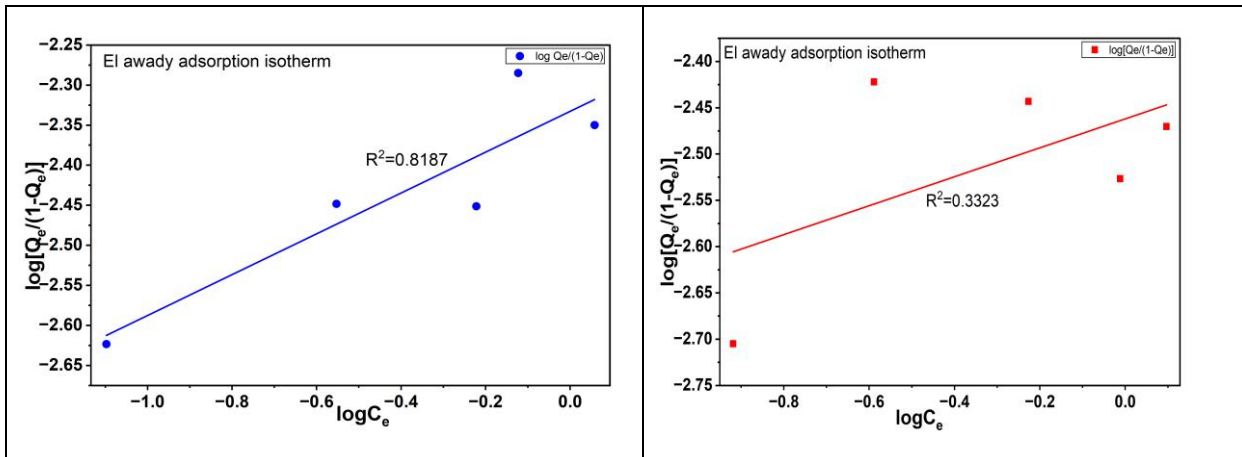


Figure 1: Various adsorption isotherms for adsorption of oxalic acid on coconut shell charcoal and bamboo charcoal

Table 3: Comparison of adsorption isotherm models with their linearized equations and corresponding R^2 values.

Adsorption isotherm	Equation for adsorption on Coconut charcoal	R^2	Equation for adsorption on Bamboo charcoal	R^2
Langmuir	$y = 202.14x + 21.657$	0.9472	$y = 300.44x + 6.7854$	0.9764
Freundlich	$y = 0.2539x - 2.3348$	0.8189	$y = 0.1559x - 2.4635$	0.3325
Temkin	$y = 0.0009x + 0.0046$	0.7465	$y = 0.0004x + 0.0034$	0.2621
Flory-Huggins	$y = 579.68x - 1.0841$	0.6007	$y = 326.23x - 1.7406$	0.0771
Frumkin	$y = -581.04x - 2.4918$	0.6002	$y = -326.42x - 4.0059$	0.0768
El-Awady	$y = 0.2548x - 2.3328$	0.8187	$y = 0.1563x - 2.462$	0.3323

The linear plots of C_e/Q_e against C_e for both the adsorbents showed good linearity, with R^2 values of 0.9472 and 0.9764 for coconut shell and bamboo charcoal, respectively, indicating that adsorption follows the Langmuir model. K_L , Q_{max} and ΔG_{ads}^0 for each were calculated using Langmuir adsorption isotherm and are summarized in the Table 4.

Table 4: Langmuir adsorption parameters (Q_{max} , K_L , and ΔG_{ads}^0) for oxalic acid on coconut shell and bamboo charcoal derived from the linearized Langmuir plots.

Adsorbent	Equation	Q_{max} (g mg ⁻¹)	K_L (L mg ⁻¹)	ΔG_{ads}^0
Coconut shell charcoal	$y=202.14x+21.657$	0.00494	9.33	-15.48
Bamboo charcoal	$y=300.44x+6.7854$	0.00333	44.27	-19.34

Langmuir parameters indicated that coconut shell charcoal had a higher Q_{max} , showing its greater ability to adsorb larger amounts of oxalic acid per gram of adsorbent. Inversely, bamboo charcoal presented a higher langmuir constant and stronger correlation, indicating a higher affinity between the oxalic acid molecules and the adsorbent surface, with more homogeneous active sites [39].

The higher value of K_L for bamboo charcoal was determined by its better surface properties, such as higher porosity, greater surface area, and plenty of oxygen functional groups, which helped in the strong hydrogen bonding and electrostatic interactions with oxalic acid [40, 41]. Therefore, although coconut charcoal had a higher adsorption capacity, bamboo charcoal presented a higher binding efficiency with stronger interaction forces.

Generally, these adsorption behaviors determined that the monolayer adsorption of oxalic acid was more effective and stable onto bamboo charcoal, proving it to be a better adsorbent for the oxalate removal process under the given conditions [42].

From K_L , ΔG_{ads}^0 i.e., Standard Gibb's free energy adsorption can be calculated by using equation (10):

$$\Delta G_{ads}^0 = -RT \ln (55.5 K_{ads}) \quad (10)$$

Within this equation, the numerical constant 55.5 denotes the molar concentration of water in the medium, arising from its density and molar mass, and is expressed in mol L⁻¹ [43].

The ΔG_{ads}^0 values for oxalic acid adsorption on coconut shell and bamboo charcoal were found to be $-15.48 \text{ kJ mol}^{-1}$ and $-19.34 \text{ kJ mol}^{-1}$, respectively. The negative values confirm that adsorption on both charcoals is spontaneous and thermodynamically favorable. Since the ΔG_{ads}^0 values lie between -20 and 0 kJ mol^{-1} , the process is predominantly physisorption [44], involving weak electrostatic and hydrogen-bonding interactions rather than chemical bonding.

The more negative value of ΔG_{ads}^0 for bamboo charcoal suggests a stronger and more favorable adsorption compared to coconut shell charcoal, possibly due to a higher surface area and porosity with an abundance of oxygen-containing functional groups. Therefore, the use of bamboo charcoal presents an effective surface for the adsorption of oxalic acid. These thermodynamic results support the conclusions from the Langmuir analysis that the adsorption of bamboo charcoal is much stronger and more spontaneous than that of coconut shell charcoal because of higher (K_L) and more negative ΔG_{ads}^0 .

IV CONCLUSIONS

The study showed effective adsorption of oxalic acid onto coconut shell and bamboo charcoal; both types of adsorption followed the Langmuir adsorption isotherm, confirming monolayer adsorption on homogeneous surface sites. Of the two adsorbents, bamboo charcoal showed a higher correlation coefficient, $R^2 = 0.9764$, and a higher Langmuir constant, K_L , revealing stronger adsorbate–adsorbent interactions and an increased surface affinity compared to coconut shell charcoal. The negative values of ΔG_{ads}^0 for the two adsorbents confirm that the adsorption is spontaneous and thermodynamically favorable. Bamboo charcoal $-19.34 \text{ kJ mol}^{-1}$ was more energetically favorable in its adsorption process than coconut shell charcoal with a $-15.48 \text{ kJ mol}^{-1}$. The magnitude of the ΔG_{ads}^0 values suggests that the adsorption proceeds mainly via physisorption by means of weak electrostatic and hydrogen-bonding interactions and not by chemical bonding. These results offer great possibilities for environmentally friendly and inexpensive biochar materials obtained from agricultural waste in the removal of oxalate pollutants from industrial and electronic waste effluents. Applying these eco-friendly adsorbents not only helps in environmental protection and resource recovery but also goes hand in hand with green chemistry and circular economy goals. Future work on surface modification or activation of these carbon materials can increase their adsorption capacity even more, standing as a promising candidate for full-scale wastewater treatment and other applications in environmental remediation.

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