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Bio-based activated carbon from coffee husk for efficient removal of Fe^{3+} and Cu^{2+} ions from water

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ABSTRACT

Contamination of water with iron and copper ions poses significant environmental and health challenges, necessitating the development of effective, sustainable treatment methods. In this study, bio-based activated carbon was synthesized from coffee husk waste through a controlled two-step thermal activation process to remove Fe^{3+} and Cu^{2+} ions from aqueous solutions. Structural characterization indicated an amorphous, mesoporous carbon material with abundant oxygen-containing functional groups and a point of zero charge of 9.25, which facilitated metal ion adsorption. Batch adsorption experiments demonstrated high removal efficiencies, and the Langmuir isotherm model predicted monolayer adsorption with maximum capacities of 23.34 mg g^{-1} for Fe^{3+} and 26.03 mg g^{-1} for Cu^{2+}. High correlation coefficients suggest uniform adsorption sites and the strong affinity of metal ions for the activated carbon, with chemisorption identified as the dominant mechanism. These findings indicate that activated carbon derived from coffee husk is a cost-effective, sustainable, and efficient adsorbent for the removal of iron and copper, supporting agricultural waste valorization and environmentally friendly water treatment.

Introduction

Contamination of water resources by heavy metal ions is a significant environmental and public health concern, primarily driven by rapid industrialization, urban expansion, and intensified agricultural activities [1]. Iron (Fe^{2+}/Fe^{3+}) and copper (Cu^{2+}) are among the most frequently detected heavy metals in surface water, groundwater, and industrial effluents, originating from sources such as mining, metal finishing, electroplating, fertilizer production, and pipeline corrosion [2,3]. While iron and copper are essential trace elements, excessive concentrations in water can lead to severe aesthetic, ecological, and health issues, including unpleasant taste and color, scaling of distribution systems, toxicity to aquatic organisms, and adverse human health effects such as gastrointestinal disorders and liver damage [4]. Conventional water treatment processes often fail to remove dissolved metal ions effectively, particularly at low concentrations, highlighting the need for more efficient and adaptable remediation technologies. Treatment methods such as chemical precipitation, ion exchange, membrane filtration, electrochemical techniques, and adsorption have been developed for heavy metal removal [5-7]. Among these, adsorption is particularly

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notable for its operational simplicity, high efficiency, low energy requirements, and minimal production of secondary pollutants. The success of adsorption processes depends heavily on the adsorbent's characteristics, prompting ongoing research into the development of cost-effective, high-performance materials capable of selectively capturing metal ions from aqueous environments.

Activated carbon is widely used for water purification because of its high surface area, well-developed porosity, chemical stability, and strong affinity toward a wide range of contaminants, including heavy metals and organic pollutants [8]. However, most commercial activated carbons are produced from non-renewable precursors such as coal and petroleum-based materials, leading to high production costs and sustainability concerns. Recently, the conversion of renewable biomass and agricultural residues into activated carbon has attracted increasing attention as a sustainable alternative that supports the circular economy and waste valorization [9]. Biomass-derived activated carbons reduce dependence on fossil resources and allow tailoring of surface chemistry and pore structure through controlled activation processes. Agricultural by-products such as rice husk, coconut shell, sugarcane bagasse, sawdust, and corn cobs have been successfully used for the adsorption of metal ions [10-14]. The removal of Fe^{3+} and Cu^{2+} ions typically involves electrostatic attraction, surface complexation with oxygen-containing functional groups, ion exchange, and pore diffusion, highlighting the importance of selecting biomass precursors with high carbon content and suitable chemical characteristics.

Coffee husk is a common byproduct of coffee processing, especially in major coffee-producing countries like Vietnam. Because it is rich in certain plant fibers, it can be used to make carbon materials with many tiny holes when heated under controlled conditions. Coffee husk also contains chemical groups that can bind to metal ions, and these can be retained or enhanced during processing. While coffee husk-based activated carbon has been widely studied for removing dyes and other pollutants, there are few detailed studies on how well it removes iron and copper, including how much it can adsorb, how quickly it adsorbs, and how it adsorbs. This study examines how well coffee husk-based activated carbon removes iron and copper from water, using tests and models to determine whether it can be a cost-effective means of removing heavy metals.

Experimental

Chemicals and Materials

Coffee husk was sourced from a local coffee processing facility, thoroughly washed with deionized water to remove impurities, and subsequently dried at 105°C for 24 hours. Stock solutions of 1000 mg L^{-1} were prepared from $FeCl_3{\cdot}6H_2O$ and $CuCl_2{\cdot}2H_2O$, respectively, using deionized water. Freshly prepared working solutions were obtained through dilution. Hydrochloric acid (HCl) and sodium hydroxide (NaOH) were used to adjust the pH. All chemicals used were of analytical grade.

Fabrication of Activated Carbon from Coffee Husk

A special oven was used to heat dried coffee husk for 2 hours at 450°C, increasing the temperature by 10°C per minute, while a steady flow of non-reactive gas was maintained. After cooling, the resulting material was crushed and sifted to produce pieces about 0.1 mm in size. Then, this material was heated again for one hour at 900°C, with the same heating rate and gas flow. After cooling in an inert gas (N_2 gas), the activated carbon was stored in sealed containers for later testing.

Characterization of activated carbon

SEM and EDX were used to assess elemental composition based on surface morphology. The phase structure was analyzed by X-ray diffraction (XRD) using Cu Kα radiation. Using Fourier transform infrared spectroscopy (4000-400 cm^{-1}), surface functional groups were identified. Textural parameters, such as specific surface area, pore volume, and pore size distribution, were evaluated using the Brunauer-Emmett-Teller (BET) method during N_2 adsorption-desorption at 77 K. The pH drift method was used to determine the point of zero charge (pH_{pzc}).

Fe^{3+} and Cu^{2+} adsorption on activated carbon

Batch adsorption tests were performed in 100 mL flasks containing 50 mL of iron or copper solution at a set initial concentration. A measured amount of AC was added, and the mixture was stirred at 150 rpm at room temperature. Samples were collected and filtered through a 0.45-micrometer filter at specific times. ICAP Q ICP-MS (Thermo Scientific, Germany) was used to quantify the residual metal. Tests examined factors such as contact time, initial metal concentration, solution pH, and AC usage. Equilibrium isotherm experiments were conducted by varying the initial Fe^{3+} or Cu^{2+} concentration while maintaining a constant AC dosage, a contact time sufficient to reach equilibrium, and a steady temperature. The equilibrium data were fitted to the Langmuir, Freundlich, Temkin, Dubinin-Radushkevich, Elovich, and Henry models.

Results and discussion

Characterization of activated carbon

SEM images of activated carbon made from coffee husks at different magnifications show a highly uneven, mixed surface (Figure 1). Satisfactory carbonization is characterized by broken, flaky carbon pieces with rough surfaces at low zoom (Fig. 1a). At higher zoom (Fig. 1b), cracks, tiny holes, and loosely stacked layers are evident, creating a structure full of pores. This surface offers numerous open sites and pathways for metal ions to bind with water.

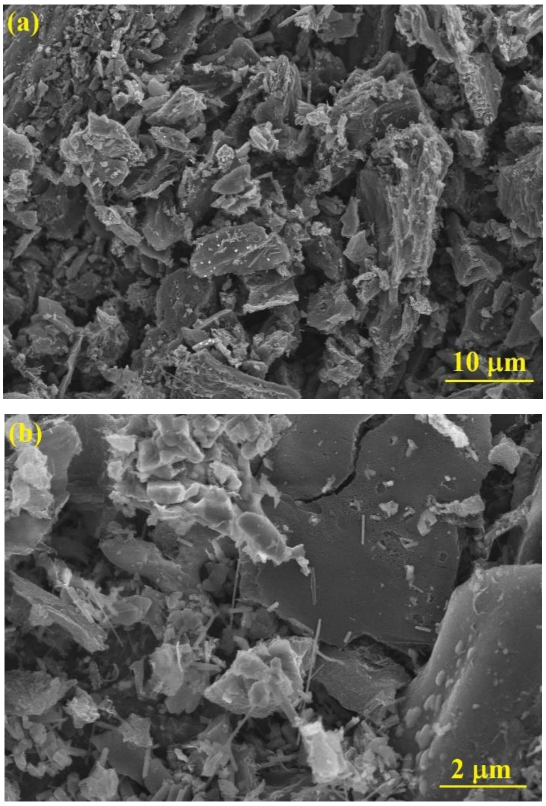
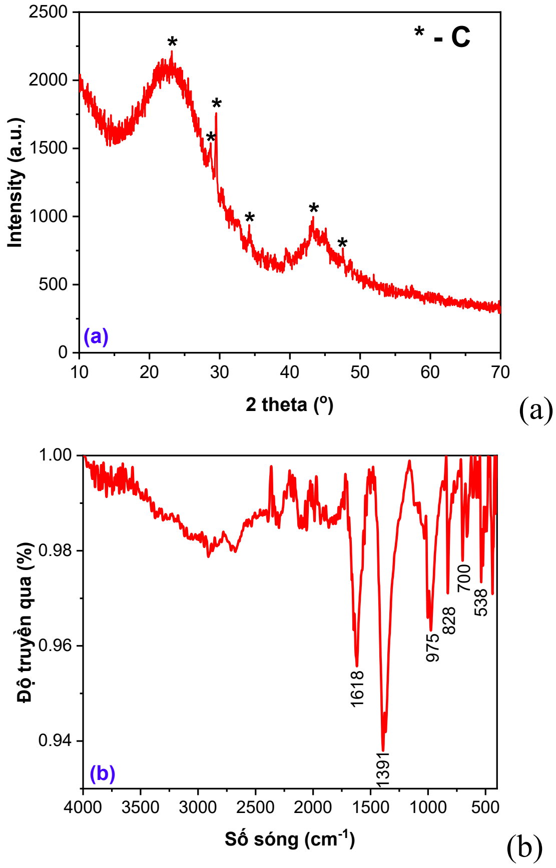


Figure 1: SEM images of coffee husk-derived activated carbon.

Figure 2a presents the XRD pattern of activated carbon derived from coffee husk, which exhibits broad diffraction peaks centered at approximately 2θ ≈ 23° and 43° [15,16]. These peaks are characteristic of amorphous carbon and correspond to the (002) and (100) planes. The absence of distinct crystalline peaks indicates the development of a disordered carbon structure during high-temperature activation, which is beneficial for adsorption applications. The FTIR spectrum (Fig. 2b) shows several functional groups containing oxygen. Bands at 1618 cm^{-1} are attributed to C=C or C=O stretching, 1391 cm^{-1} to C-H bending or carboxylate groups, and peaks at 975, 828, and 700 cm^{-1} are associated with C-O stretching and aromatic C-H vibrations [17]. The presence of these functional groups enhances metal ion adsorption via surface complexation and electrostatic interactions.

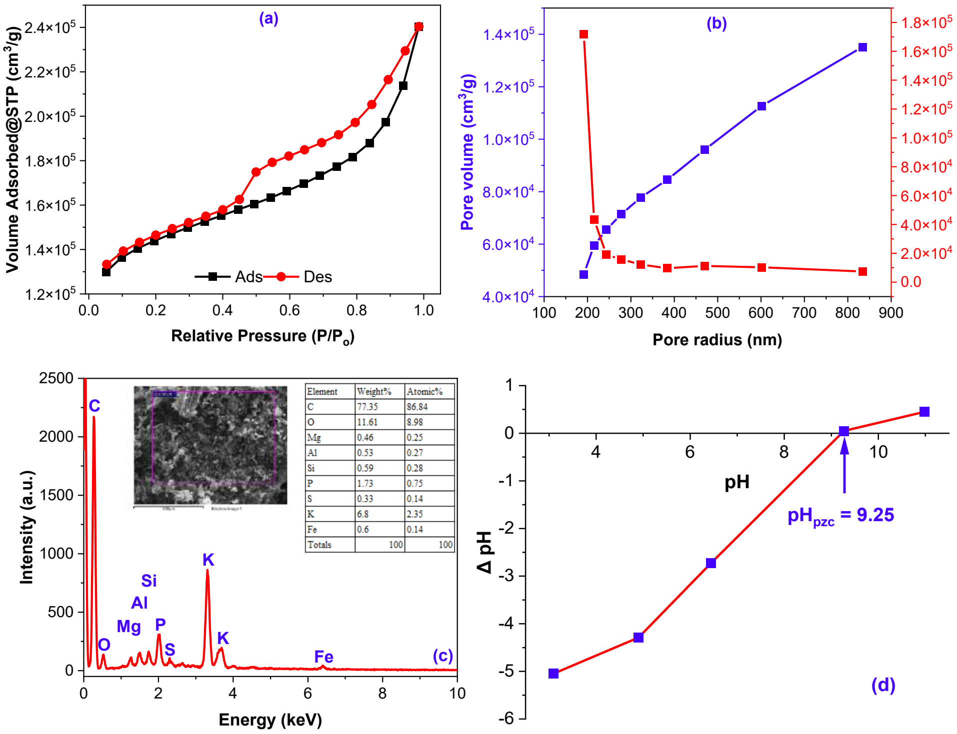


(a)

(b)

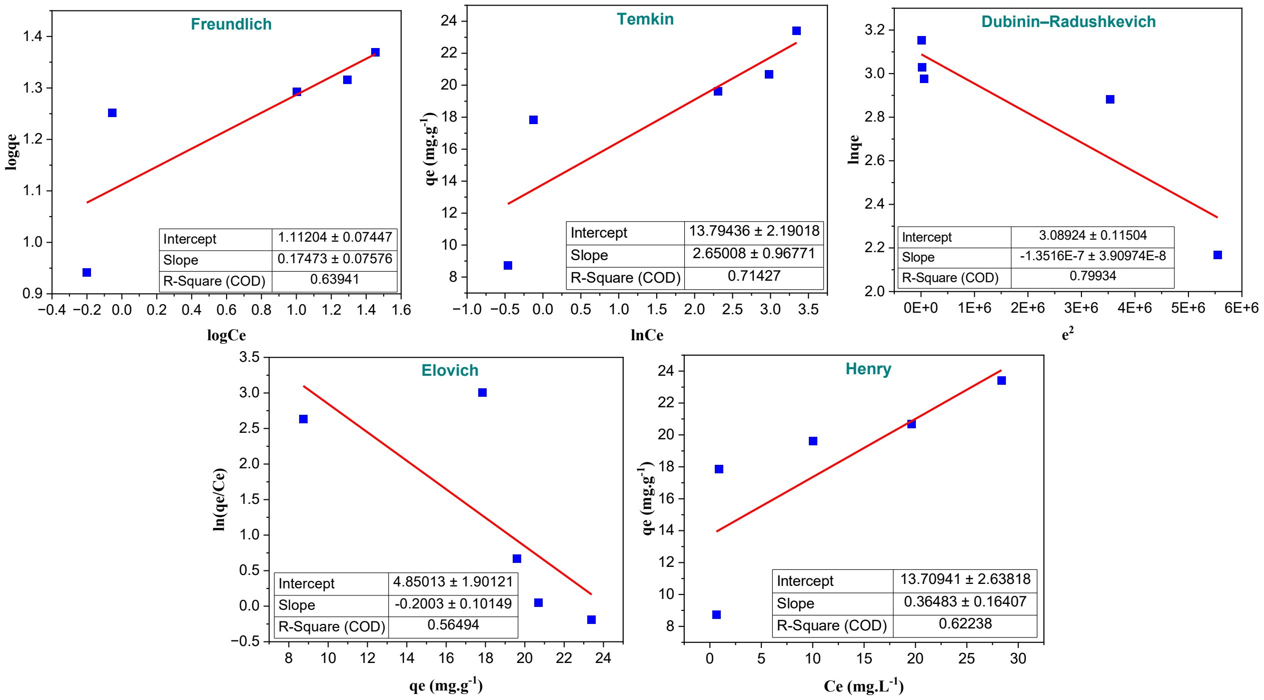
Figure 2: XRD pattern (a) and FTIR spectroscopy (b) of coffee husk-derived activated carbon.

Figure 3a shows the N_2 adsorption-desorption isotherm of coffee husk-derived activated carbon, which exhibits a typical type IV profile with a hysteresis loop, indicating a mesoporous structure. The BJH pore size distribution (Fig. 3b) further confirms the dominance of mesopores with a broad pore size range, favorable for metal ion diffusion and adsorption. The EDX pattern (Fig. 3c) reveals that carbon is the main constituent, accompanied by oxygen and minor elements such as K, Si, Mg, Al, and Fe, originating from the biomass precursor and contributing potential active sites. Figure 3d shows the point of zero charge (pH_pzc) of 9.25 indicates that the material surface is positively charged at pH values below 9.25 and becomes negatively charged when the pH exceeds this value. Since Fe^{3+} and Cu^{2+} exist as cationic species in aqueous solution, electrostatic attraction is thermodynamically more favorable at higher pH, where the surface acquires a negative charge. However, under practical water treatment conditions, effective removal can still be achieved at near-neutral pH through additional mechanisms, such as surface complexation with oxygen-containing functional groups or ion-exchange interactions. Therefore, an appropriate operational pH range should balance enhanced adsorption performance with the need to avoid metal hydroxide precipitation, which typically occurs at elevated pH values for Fe^{3+} and Cu^{2+}.



Element	Weight%	Atomic%
C	77.35	86.84
O	11.61	8.98
Mg	0.46	0.25
Al	0.53	0.27
Si	0.59	0.28
P	1.73	0.75
S	0.33	0.14
K	6.8	2.35
Fe	0.6	0.14
Totals	100	100

Figure 3: N_2 adsorption-desorption isotherm (a), BJH pore size distribution (b), EDX pattern (c), and pH_{pzc} (d) of coffee husk-derived activated carbon.



Freundlich

Intercept	1.11204 ± 0.07447
Slope	0.17473 ± 0.07576
R-Square (COD)	0.63941

Temkin

Intercept	13.79436 ± 2.19018
Slope	2.65008 ± 0.96771
R-Square (COD)	0.71427

Dubinin–Radushkevich

Intercept	3.08924 ± 0.11504
Slope	-1.3516E-7 ± 3.90974E-8
R-Square (COD)	0.79934

Elovich

Intercept	4.85013 ± 1.90121
Slope	-0.2003 ± 0.10149
R-Square (COD)	0.56494

Henry

Intercept	13.70941 ± 2.63818
Slope	0.36483 ± 0.16407
R-Square (COD)	0.62238

Figure 4: Fe^{3+} adsorption isotherm models on coffee husk-derived activated carbon at room temperature for 8 hours

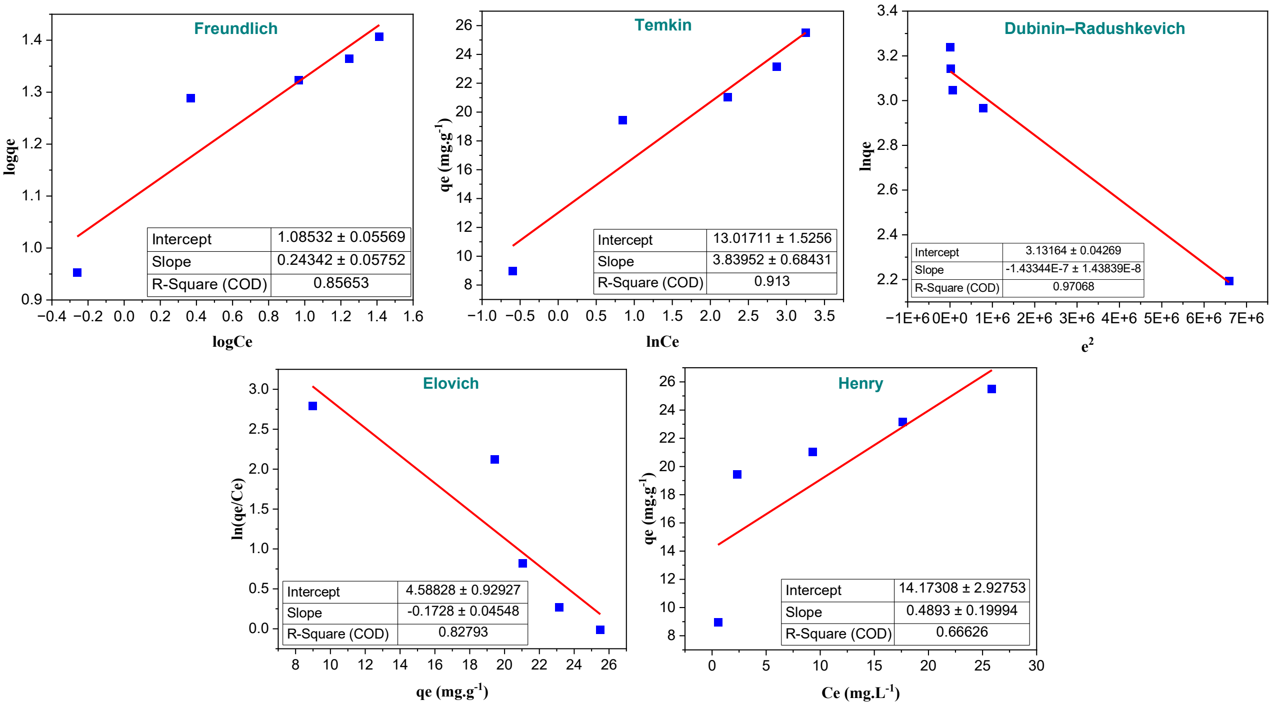
Fe^{3+} and Cu^{2+} removal by activated carbon

Figure 4 and Table 1 illustrate the adsorption isotherms of Fe^{3+} on coffee husk-derived activated carbon. Among the evaluated models, the Langmuir isotherm shows the best fit (R^2 = 0.99191), indicating monolayer adsorption on a homogeneous surface with a maximum adsorption capacity of 23.34 mg g^{-1}, reflecting strong affinity between Fe^{3+} ions and the carbon surface. The Freundlich, Temkin, Henry, and Elovich models exhibit lower correlation coefficients, suggesting limited applicability in describing the adsorption behavior. The Dubinin-Radushkevich model indicates significant adsorption energy, implying that chemisorption contributes to Fe^{3+} uptake. Overall, the results confirm that Fe^{3+} adsorption on coffee husk-derived activated carbon is predominantly governed by monolayer adsorption with strong surface interactions.

Figure 5 and Table 1 show that Cu^{2+} adsorption on coffee husk-derived activated carbon is best described by the Langmuir model (R^2 = 0.99453), indicating monolayer adsorption on a relatively homogeneous surface with a maximum capacity of 26.03 mg g^{-1} and strong affinity (K_L = 0.74027 L mg^{-1}). The Freundlich and Henry models show lower correlations, suggesting limited heterogeneity and linear adsorption behavior. The Temkin and Dubinin-Radushkevich models provide moderate fits, implying adsorbate-adsorbent interactions and an essential contribution of chemisorption. Overall, Langmuir dominance confirms efficient and uniform Cu^{2+} uptake by the activated carbon surface.

Table 1: Parameters of Fe^{3+} and Cu^{2+} adsorption isotherm models on activated carbon

Models	Parameter	Value	
		Fe^{3+}	Cu^{2+}
Langmuir	K_L ($L.mg^{-1}$)	0.877	0.740
	q_{max} ($mg.g^{-1}$)	23.343	26.028
	R^2	**0.9919**	**0.9945**
Freundlich	K_F $(mg.g^{-1}).(L.mg^{-1})^{1/n}$	12.943	12.171
	n	5.723	4.108
	R^2	0.639	0.857
Temkin	K_T	182.229	29.675
	b_T ($kJ.mol^{-1}$)	2.650	3.839
	R^2	**0.7143**	**0.9130**
Dubinin-Radushkevich	q_{max} ($mg.g^{-1}$)	21.960	22.912
	β ($mol^2.kJ^{-2}$)	$1.351.10^{-7}$	$1.433.10^{-7}$
	E ($J.mol^{-1}$)	1,923.362	1,867.649
	R^2	0.7993	0.9707
Elovich	α ($mg.g^{-1}$)	127.757	98.325
	β ($g.mg^{-1}$)	0.200	0.173
	R^2	0.565	0.828
Henry	K_H	0.365	0.489
	R^2	0.622	0.666



Freundlich	
Intercept	1.08532 ± 0.05569
Slope	0.24342 ± 0.05752
R-Square (COD)	0.85653

Temkin	
Intercept	13.01711 ± 1.5256
Slope	3.83952 ± 0.68431
R-Square (COD)	0.913

Dubinin–Radushkevich	
Intercept	3.13164 ± 0.04269
Slope	-1.43344E-7 ± 1.43839E-8
R-Square (COD)	0.97068

Elovich	
Intercept	4.58828 ± 0.92927
Slope	-0.1728 ± 0.04548
R-Square (COD)	0.82793

Henry	
Intercept	14.17308 ± 2.92753
Slope	0.4893 ± 0.19994
R-Square (COD)	0.66626

Figure 5: Cu^{2+} adsorption isotherm models on coffee husk-derived activated carbon at room temperature for 8 hours

Conclusion

Coffee husk-derived bio-based activated carbon efficiently and sustainably removes Fe^{3+} and Cu^{2+} ions from aqueous systems. A two-step thermal activation process produces a mesoporous, amorphous carbon material with abundant oxygen-containing functional groups and a favorable surface charge. The Langmuir model fits the equilibrium data best. This means that the carbon surface has a strong affinity for metal ions and can only hold one layer of them. Adsorption measurements confirm substantial metal uptake. Maximum adsorption capacities of 23.34 mg g^{-1} for Fe^{3+} and 26.03 mg g^{-1} for Cu^{2+} highlight the material's competitiveness. This approach offers a cost-effective method for heavy metal remediation while supporting the valorization of agricultural waste and sustainable water treatment.

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