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SULFATED FUCOIDAN HYDROGEL FROM *Undaria pinnatifida* AS A HIGH-PERFORMANCE BIOADSORBENT FOR CADMIUM REMOVAL FROM WASTEWATER

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ABSTRACT

Cadmium (Cd^{2+}) must be effectively removed from effluent because of its high toxicity and persistence in aquatic settings. In this work, *Undaria pinnatifida* was used to create a sulfated fucoidan hydrogel (FUC-HG), which was then tested as a high-performance bioadsorbent for Cd^{2+} sequestration. A maximal adsorption capacity of approximately 50 mg g^{-1} was made possible by the hydrogel's heterogeneous surface with numerous active binding sites. While kinetic analysis followed the pseudo-second-order model (R^2=0.9876), confirming chemisorption as the main mechanism, equilibrium data fitted the Freundlich isotherm (R^2=0.9441), showing multilayer adsorption. According to thermodynamic investigations, the reaction was spontaneous, endothermic, and entropy-driven, which is in line with chemical interactions between the hydroxyl and sulfated functional groups of FUC-HG and Cd^{2+} ions. Elovich and intraparticle diffusion investigations, which emphasised surface binding followed by pore diffusion, provided additional evidence for sequential uptake. Over several cycles, the hydrogel showed outstanding durability and reusability while retaining a high adsorption efficiency. These findings establish FUC-HG as an economical, selective, and eco-friendly bioadsorbent with great promise for real-world use in industrial heavy metal remediation and wastewater treatment.

1. Introduction

One of the most serious environmental problems brought on by fast industrialisation, mining, electroplating, battery manufacturing, and fertiliser production is heavy metal poisoning of aquatic systems [1]. Cadmium (Cd^{2+}) is one of the most dangerous metals because of its high water mobility, lengthy biological half-life, and extreme toxicity even at low levels [2]. International regulatory bodies have imposed stringent discharge limitations on cadmium due to its association with kidney dysfunction, skeletal damage, carcinogenicity, and ecological bioaccumulation [3]. Nonetheless, a major scientific and technological challenge remains in effectively removing Cd^{2+} from complicated effluent streams under practical conditions.

Chemical precipitation, ion exchange resins, membrane filtering, and electrochemical treatments are examples of conventional cadmium removal methods that frequently have disadvantages such as high operating costs, secondary sludge production, limited selectivity, and poor regeneration efficiency [4] . Because of their ease of use, scalability, and ability to adjust to low metal concentrations, adsorption-based methods have become one of the most promising substitutes. However, a lot of reported adsorbents rely on inorganic materials, synthetic polymers, or activated carbons that are not sustainable, need energy-intensive manufacturing processes, or function poorly in changeable wastewater conditions. Growing interest in bio-based, renewable, and ecologically safe adsorbents that can achieve high adsorption efficiency without sacrificing ecological safety has resulted from this [5-16].

Because of their abundance, rich surface chemistry, and intrinsic biodegradability, marine polysaccharides have garnered significant interest as next-generation adsorption materials [17-26]. Fucoidan, a sulfated polysaccharide derived from brown seaweeds like Undaria pinnatifida, is especially appealing due to its high density of hydroxyl (–OH) and sulphate ($–SO_3^-$) functional group. Fucoidan is a viable option for selective heavy metal binding because of these properties, which offer strong electrostatic interactions and coordination sites for divalent metal ions [27]. However, natural fucoidan's water solubility, poor mechanical stability, and challenges with recovery following adsorption limit its practical usage in wastewater treatment systems.

Fucoidan's structural alteration into a crosslinked hydrogel (FUC-HG) provides a logical way to improve its physicochemical stability while maintaining its inherent metal-binding capability in order to get over these restrictions [28]. Rapid mass transfer, fast water uptake, and active site accessibility are all made possible by hydrogels' three-dimensional porous network. These hydrogels are excellent for aqueous metal remediation because they combine ion-exchange capacity, chemical affinity, and structural robustness when produced from sulfated biopolymers. Despite these benefits, there are still few systematic studies in the literature on sulfated fucoidan-based hydrogels for cadmium adsorption, especially with thorough kinetic, isothermal, thermodynamic, and mechanistic evaluations.

This study generated and assessed a sulfated fucoidan hydrogel (FUC-HG) obtained from Undaria pinnatifida as a high-performance bioadsorbent for the removal of Cd^{2+} from aqueous environments. The effects of temperature, initial Cd^{2+} content, and dosage were investigated by batch adsorption studies [29]. In order to clarify the governing adsorption pathways, the adsorption behaviour was thoroughly examined using a variety of kinetic and isotherm models in addition to thermodynamic and mechanistic interpretations. This work intends to improve the use of marine biopolymer-based hydrogels in cutting-edge wastewater treatment technologies and establish FUC-HG as a practical, environmentally friendly substitute for traditional adsorbents by

combining sustainability, high adsorption efficiency, and reusability.

2. Materials and Methods

2.1. Materials

The main source of fucoidan was commercially accessible dried brown seaweed (*Undaria pinnatifida*). Sigma-Aldrich provided analytical-grade cadmium nitrate tetrahydrate ($Cd(NO_3)_2.4H_2O$), ethanol (≥99%), hydrochloric acid, sodium hydroxide, calcium chloride, acetone, and sodium chloride, all of which were utilised without additional purification. Deionised water (18.2 MΩ.cm) was used to prepare all solutions. Thermo Scientific supplied dialysis tubing with a molecular weight cut-off of 12 kDa. Analytical grade reagents were utilised for all other polysaccharide analysis and adsorption tests.

2.2. Extraction and Purification of Sulfated Fucoidan

A modified hot-water extraction process that maximises yield and preserves sulphate groups was used to extract fucoidan from *Undaria pinnatifida*. After being properly cleaned with deionised water to get rid of salts and surface contaminants, the dried seaweed was air-dried, powdered, and then defatted by refluxing in 70% ethanol to get rid of lipids and colours. The pretreatment biomass (50 g) was heated to 85 °C for three hours while being continuously stirred. It was suspended in deionised water at a material-to-solvent ratio of 1:20 (w/v). After cooling and filtering out insoluble residues, the extract was treated with 0.2 M $CaCl_2$ to preferentially precipitate alginates. Following centrifugation to remove the alginate fraction, the cleared extract was concentrated under low pressure and ethanol precipitation was applied by gradually adding cooled ethanol to reach a final concentration of 75% (v/v). Centrifugation was used to recover the precipitated polysaccharide, which was then twice cleaned with 70% ethanol and acetone, dissolved in water, and deproteinised using papain enzymatic treatment at 55 °C for four hours. After the enzyme was deactivated by heating the solution, it was dialysed against deionised water for 72 hours with frequent water changes. Purified sulfated fucoidan powder was obtained by lyophilising the dialysate.

2.3. Preparation of Fucoidan Hydrogel (FUC-HG)

Ionic crosslinking was used to create FUC-HG, a stable three-dimensional network appropriate for cadmium adsorption. A homogenous viscous solution was achieved by dissolving purified fucoidan in warm deionised water (4% w/v) and stirring the mixture. The polymer solution was added dropwise to a 2% (w/v) $CaCl_2$ solution that was gently agitated. This allowed for the instantaneous creation of gel beads, which were caused by ionic coordination between Ca^{2+} and sulphate groups. The resulting hydrogel beads were periodically cleaned with deionised water to get rid of unattached ions after being cured for 24 hours at 4 °C to improve structural stability. For batch adsorption investigations, the hydrogels were crushed to the appropriate particle size after being dried at 50 °C.

2.4. Batch Adsorption Experiments

The effectiveness of FUC-HG in removing Cd^{+2} at different physicochemical conditions was assessed using Batch Adsorption experiments. 50 mL of Cd^{2+} solutions with starting concentrations ranging from 10 to 300 mg/L were mixed with a known mass of hydrogel (usually 20–30 mg). Diluted HCl or NaOH was used to modify the pH of the solutions between 2 and 7 while the suspensions were shaken at 150 rpm in a thermostatic orbital shaker. Depending on the kinetic study design, the contact period ranged from 5 to 240 minutes, and temperature effects were investigated at 25, 35, and 45 °C. Following adsorption, the solutions were filtered, and AAS was used to measure the residual Cd^{2+} concentration. Standard mass balance equations were used to compute the removal effectiveness (%) and adsorption capacity (q_e).

2.5. Isotherm Modelling

Five popular isotherm models the Langmuir, Freundlich, Temkin, Dubinin-Radushkevich

(D-R), and Elovich models were fitted to experimental data in order to clarify the equilibrium adsorption behaviour. The maximum monolayer adsorption capacity (q_m), adsorption intensity (n), binding constant (K^1), surface heterogeneity, and mean free energy of adsorption were all determined using these models. Mechanistic interpretation of surface homogeneity, monolayer versus multilayer adsorption, and the potential energy of interaction between Cd^{2+} and sulfate-rich functional sites was made possible by the thorough comparison.

2.6. Adsorption Kinetics

Time-dependent adsorption studies at an ideal concentration of Cd^{2+} were used to assess kinetic behaviour. Five kinetic models pseudo-first-order, pseudo-second-order, Elovich, intraparticle diffusion, and Pore Diffusion models were fitted to the acquired q_t vs. time curves. Nonlinear regression was used to fit each model in order to ascertain the kinetic order, diffusion parameters, sorption intensity, and rate constants. By differentiating between film diffusion, intraparticle transport, chemisorption, and multimechanistic routes that contribute to Cd^{2+} uptake by the fucoidan hydrogel, the combination of kinetic models allows for a thorough understanding of the governing rate-limiting stages.

2.7. Thermodynamic Analysis

To comprehend the viability and energetic nature of Cd^{2+} adsorption, thermodynamic characteristics were assessed. The van't Hoff equation was utilised to compute changes in Gibbs free energy (ΔG°), enthalpy (ΔH°), and entropy (ΔS°) utilising distribution coefficients (K_e) acquired at various temperatures. The spontaneity, endo/exothermic nature, and increased unpredictability related to the adsorption process on the FUC-HG surface were shown by the sign and magnitude of these characteristics.

2.8. Reusability and Stability Tests

Adsorption-desorption cycles were used to evaluate the hydrogel's capacity for regeneration. After being desorbed with 0.1 M HCl and cleaned with deionised water until the pH was neutral, loaded hydrogels were recycled for further cycles under the same circumstances. The hydrogel's structural resilience and the strength of the Cd^{2+} binding interactions were assessed by measuring the loss in adsorption efficiency and evaluating the stability and reusability across five consecutive cycles.

3. Results and Discussion

3.1. Influence of pH on Cadmium Uptake and Surface Chemistry

The interaction between Cd^{2+} ions and the FUC-HG was significantly impacted by the pH of the solution. Protonation of sulphate groups reduced the availability of negatively charged binding sites under extremely acidic circumstances, leading to low absorption [30]. Deprotonation of $–SO_3^-$ groups boosted the electrostatic attraction towards Cd^{2+} as pH rose towards the ideal range, resulting in a dramatic increase in adsorption capacity. The creation of cadmium hydroxide species, which decreased the population of free Cd^{2+} ions available for binding, was thought to be the cause of minor drops in adsorption beyond the ideal threshold. This pH-dependent trend demonstrated the strong pH-responsive binding behaviour of the sulfate-rich fucoidan matrix and the predominance of electrostatic attraction and metal sulfate coordination in Cd^{2+} adsorption. The hydrogel's potential for practical use under environmental pH ranges characteristic of contaminated water systems was underlined by the well-defined pH window for maximal adsorption.

3.2. Effect of Adsorbent Dosage and Surface Accessibility

A progressively higher availability of active functional sites was suggested by a gradual improvement in removal efficiency with increasing hydrogel dosage. However, because of unsaturated binding sites, particle aggregation, and lower driving force per unit mass, the equilibrium adsorption capacity (q_e) dropped at higher dosages [31]. Biopolymer-based hydrogels frequently exhibit this behaviour, where high swelling capacity causes polymer domains to overlap

at high doses, limiting ion migration into deeper matrix regions [32]. The plateauing pattern indicated that the FUC-HG has both accessible surface sites and a highly porous interior structure that facilitates efficient diffusion of Cd^{2+} under optimal dose. These findings emphasise how crucial dosage control is for maximising cost effectiveness in real-world water treatment applications rather than for boosting intrinsic capability.

3.3. Effect of Initial Cadmium Concentration and Mass Transfer Driving Force

Because of the large ratio of accessible binding sites to Cd^{2+} ions, the hydrogel quickly reached equilibrium at lower starting concentrations. The adsorption curve changed from a high-affinity region to a saturation-driven plateau as concentration rose, indicating the active sulphate groups' increasing occupied. Increased mass transfer rate and quicker diffusion through the hydrogel matrix were made possible by higher driving force at higher Cd^{2+} levels. But as the hydrogel gets closer to its natural loading capacity, the final saturation showed that the adsorption mechanism changes from site-rich to site-limited circumstances. The behaviour verified the balance between intraparticle transport and exterior film diffusion, which controls the overall uptake process, as well as the strong affinity between Cd^{2+} and the fucoidan network.

3.3.1. Adsorption isotherms

The performance evaluation of any sorbent is purely based on its ability to uptake the maximum concentration of heavy metals from aqueous solutions. Therefore, the batch experimental data in Table. 1. obtained from the effect of the initial concentration of Cd(II) on sorption capacity of the sorbent was applied to the linear forms of Freundlich, Langmuir, D-R, and Elovich, and Temkin model equations to know about the maximum sorption capacity of FUC-HG and mechanism of sorption phenomenon. Also, the physisorption or chemisorption nature of the sorption process was predicted from these isotherms**.**

3.3.1.1. Freundlich Isotherm

The Freundlich isotherm analysis (log q_e vs. log C_e, R^2 = 0.9441) indicates the heterogeneous surface adsorption behavior of FUC-HG, in which several binding sites with different affinities are involved in the uptake of ions ~50 mg/g. This strong fit suggests that the hydrogel's uptake of metal ions is effectively captured by the isotherm. The Freundlich relationship is mathematically given by

$$q_e = K_F C_e^{1/n} \quad \text{(Eq. 1)}$$

in which q_e denotes the amount of solute per unit mass of adsorbent at equilibrium, C_e is the corresponding equilibrium concentration of the ions. K_F represents a capacity-related constant, and 1/n provides insight into the degree of surface site heterogeneity. The Freundlich equation in linear form is:

$$\log q_e = \log K_F + \frac{1}{n}\log C_e \quad \text{(Eq. 2)}$$

The slope (1/n = 0.7233), as seen in Fig. 1A, is between 0 and 1, indicating favourable adsorption and notable surface heterogeneity. The fucoidan network's abundance of sulfated groups ($–SO_3^-$) and remaining hydroxyl functionalities, which provide adsorptive sites with varying energy levels and capabilities for Cd^{2+} binding, are the cause of this variability. These high- affinity surface sites facilitate quick and effective uptake at lower metal ion concentrations; as concentration rises, these sites gradually get occupied, resulting in multilayer accumulation on lower-energy areas of the polymeric matrix. As a result, FUC-HG functions as a non-uniform, heterogeneous sorbent that can hold Cd^{2+} by a variety of interactions, including complexation, ion exchange, and electrostatic attraction. As a result, the FUC- HG varied chemical activity permits strong adsorption over a broad concentration range, indicating its promise as a flexible and effective bioadsorbent for wastewater treatment.

3.3.1.2. Langmuir Isotherm

The Langmuir isotherm model (C_e/q_e vs C_e, R^2 = 0.8792) confirms that it operates by

monolayer adsorption capacity with FUC-HG which absorbed metal ions from the solution to reach finite binding sites evenly distributed throughout the surface shown in Fig. 1B. The Langmuir equation is

$$q_e = \frac{Q_{\max} b C_e}{1 + b C_e} \quad \text{(Eq. 3)}$$

where q_e is the amount of adsorbate adsorbed at equilibrium (mg g^{-1}), Q_{max} is the maximum monolayer adsorption capacity (mg g^{-1}), b is the Langmuir constant related to the affinity of binding sites (L mg^{-1}), C_e is the equilibrium concentration of adsorbate in solution (mg L^{-1}). The linear form of Langmuir equation is

$$\frac{C_e}{q_e} = \frac{1}{Q_{\max} \times b} + \frac{C_e}{Q_{\max}} \quad \text{(Eq. 4)}$$

The Langmuir isotherm's positive slope and distinct intercept indicate that each adsorption site on FUC-HG holds a single Cd^{2+} ion, which is compatible with the idea of monolayer surface coverage. This behaviour suggests the existence of spatially well-defined and energetically equivalent binding sites in the hydrogel matrix, which are mainly linked to the hydroxyl and sulfated ($–SO_3^-$) functional groups present in the fucoidan backbone. Through ion-exchange interactions and electrostatic attraction, these functional moieties offer particular coordination conditions that promote selective Cd^{2+} binding.

Although the Freundlich model emphasises surface heterogeneity and multilayer adsorption behaviour, the Langmuir model's strong applicability shows that uniform, site-specific interactions account for a sizable portion of Cd^{2+} uptake. A dual adsorption mechanism in FUC-HG, comprising both monolayer adsorption on high-energy, homogeneous sites and multilayer accumulation on heterogeneous portions of the hydrogel network, is indicated by the coexistence of Freundlich and Langmuir properties. By expanding the operational concentration range and improving ion-exchange efficiency, this coupled adsorption behaviour allows FUC-HG to remove Cd^{2+} ions from aqueous environments in a highly effective and selective manner.

3.3.1.3. Dubinin–Radushkevich (D–R) isotherm

From the D–R isotherm plot (ln q_e vs. ε^2, R^2 = 0.9878) in Fig. 1C. it can be understood that the adsorption of heavy metal ions by FUC-HG is conditioned best by this model and a chemisorption interception controlled process takes place. The D-R equation is given by

$$q_e = X_m e^{-\beta \varepsilon^2} \quad \text{(Eq. 5)}$$

Where q_e is the equilibrium adsorption capacity (mg g^{-1}), X_m is the theoretical saturation capacity (mg g^{-1}), β is the D-R constant ($mol^2\ J^{-2}$), related to adsorption energy, ε is the Polanyi potential calculated through $\varepsilon = RT \ln(1+1/C_e)$. The linear form of D-R equation is

$$\ln q_e = \ln X_m - \beta \varepsilon^2 \quad \text{(Eq. 6)}$$

$$\varepsilon^2 = RT \ln\left(1 + \frac{1}{C_e}\right) \quad \text{(Eq. 7)}$$

$$E = \frac{1}{\sqrt{-2\beta}} \quad \text{(Eq. 8)}$$

The computed mean adsorption energy (E > 8 kJ mol^{-1}) suggests that chemical adsorption, rather than weak physical interactions like van der Waals forces [32], is the primary mechanism of Cd^{2+} uptake by FUC-HG. The presence of high-affinity binding sites on the hydrogel surface that interact with Cd^{2+} ions selectively is confirmed by the size of the adsorption energy.

The predominance of chemisorption indicates that ion-exchange and coordination mechanisms, wherein sulfated ($–SO_3^-$) and hydroxyl (–OH) functional groups within the fucoidan backbone create strong ionic or coordination interactions with Cd^{2+} ions, are the main mechanisms by which FUC-HG operates. The metal ions in the hydrogel network are stabilised as a result of these interactions. The high adsorption efficiency,

selectivity, and specificity of FUC-HG towards Cd^{2+} are explained by the frequency of such chemically driven interactions, highlighting its great promise as an efficient bioadsorbent for the removal of hazardous cadmium ions from contaminated wastewater.

3.3.1.4. Elovich Model

The adsorption behaviour of Cd^{2+} ions onto FUC-HG can be better understood using the Elovich kinetic model (Fig. 1D), especially in situations where chemically controlled processes predominate. Because the fucoidan hydrogel has a large number of sulfated ($-SO_3^-$) and hydroxyl (–OH) functional groups dispersed throughout the polymeric network, this model is well suited for explaining adsorption on energetically heterogeneous surfaces. Strong chemisorptive binding rather than straightforward physical adsorption is the outcome of these multifunctional sites' active participation in ion-exchange and coordination interactions with Cd^{2+} ions.

The linear form of Elovich equation is

$$\ln\frac{q_e}{C_e} = \ln K_E Q_{\max} - \frac{q_e}{Q_{\max}} \qquad \text{(Eq. 9)}$$

A dual-stage adsorption mechanism for Cd^{2+} uptake is suggested by the Elovich kinetic behaviour of FUC-HG. Due to the large number of easily accessible high-energy binding sites on the hydrogel surface, where Cd^{2+} ions are swiftly immobilised via monolayer ion-exchange and coordination contacts, adsorption proceeds quickly in the first stage [33]. This rapid uptake is a result of the fucoidan network's sulfated and hydroxyl functional groups having a significant affinity for Cd^{2+} ions and low surface coverage.

Diffusion-controlled multilayer adsorption within the porous hydrogel matrix dominates the second stage, which is indicated by the rate steadily declining as adsorption advances. During this stage, Cd^{2+} ions go into the interior of FUC-HG and engage in stronger chemisorption interactions with functional groups. The heterogeneous and chemically controlled nature of Cd^{2+} adsorption is highlighted by the coexistence of rapid surface binding and slower intraparticle diffusion, indicating that FUC-HG functions effectively over a broad concentration range a crucial characteristic for realistic wastewater treatment applications [34].

3.3.1.5. Temkin Isotherm

The Temkin isotherm characterizes systems where the interaction energy between solute and substrate diminishes uniformly as surface coverage rises, thereby highlighting the importance of solute - substrate cooperative interactions [35]. The linear q_e versus ln C_e relationship observed for FUC-HG ($R^2 = 0.9682$) corroborates this model and indicates that solute–substrate interactions materially influence the uptake. The mathematical formulation is given by

$$q_e = \frac{RT}{b}\ln(K_T C_e) \qquad \text{(Eq. 10)}$$

where R is the universal gas constant, T is absolute temperature, K_t is the Temkin affinity constant, and b is linked to the adsorption heat. The linear form of temkin equation is

$$q_e = B\ln A + B\ln C_e \qquad \text{(Eq. 11)}$$

The effective heat of adsorption during Cd^{2+} absorption by FUC-HG is reflected in the rising trend of the q_e versus ln C_e plot (Fig. 1E). Due to the sequential occupation of high-energy binding sites and electrostatic repulsion among adsorbed ions, this behaviour suggests that the adsorption energy gradually reduces with increasing Cd^{2+} concentration. Cd^{2+} ions first preferentially occupy energetically favourable sulfate-rich locations on the hydrogel surface; as these sites get saturated, adsorption moves to lower-energy areas of the hydrogel matrix, which lowers the incremental adsorption strength.

This kind of behaviour is typical of sulfated biopolymer hydrogels, where coordination with functional groups, ion-exchange interactions, and electrostatic attraction all

work together to control Cd^{2+} binding instead of just weak physical forces [35]. The success of FUC-HG as a high-performance ion-exchange material for cadmium removal from aqueous systems is confirmed by the progressive filling of sites with decreasing adsorption energy, which supports a multilayer adsorption mechanism

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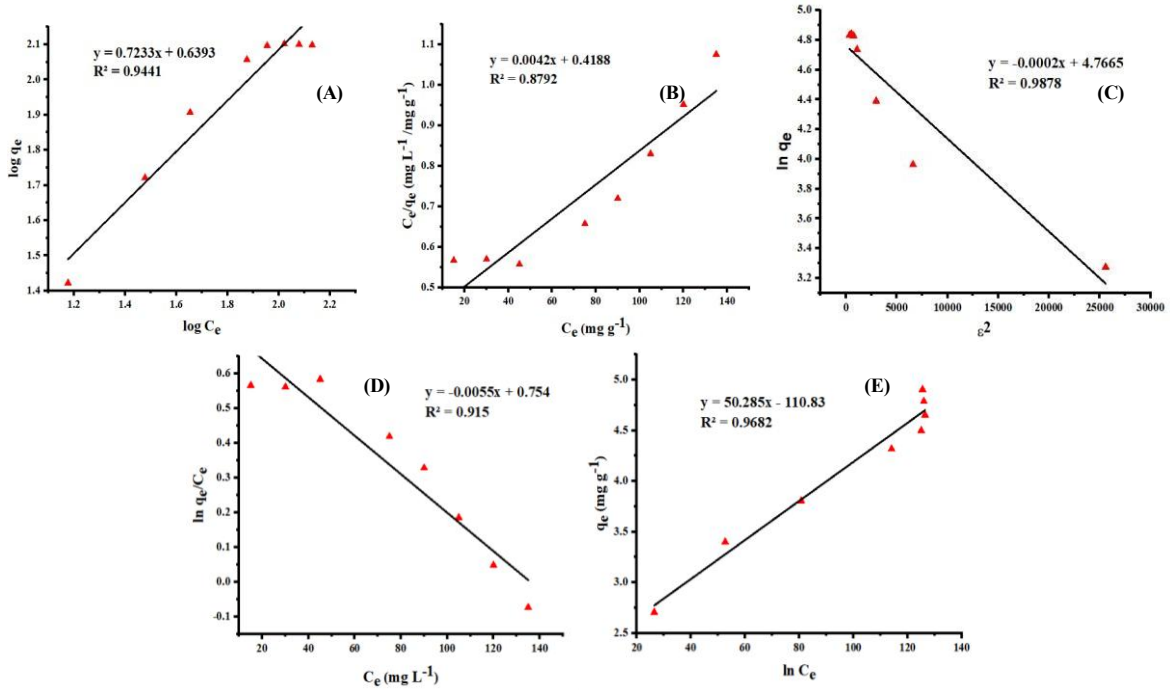

Fig. 1. Adsorption isotherms of Cd^{+2} onto FUC-HG. (A) Freundlich Isotherm, (B) Langmuir Isotherm, (C) Dubinin–Radushkevich Isotherm, (D) Elovich Model, and (E) Temkin Isotherm

Table. 1. The isothermal parameters of linear forms of Freundlich, Langmuir, Dubinin-Radushkevich, Elovich, and Temkin models for the Cd^{+2} uptake by FUC-HG.

Experimental	q_e (mgg^{-1})	126.4381
Freundlich	K_F	4.358127
	N	1.382607
	R^2	0.9441
Langmuir	Q_{max} (mg g^{-1})	238.2801
	b (mg L^{-1})	0.010021
	R_L	2.352858
	R^2	0.8792
D-R	$X_m=Q_{max}$ (mg g^{-1})	116.9396
	β (kJ^2/mol^2)	-2.52201E-08
	E	4452.574
	R^2	0.9878
Elovich	Q_{max} (mg g^{-1})	201.8114
	KE(Lmg^{-1})	1.00002
	R^2	0.915
Temkin	B	50.2847
	A	0.110347
	$b_T(J\ mol^{-1})$	49.27089
	R^2	0.9682

3.3.2. Kinetic Models

The appraisal of sorption kinetics is one of the key factors to evaluate the rate at which sorbent can uptake the highest amounts of heavy metals from the parent contaminated systems. The kinetic study of the sorption process gives direct insight into reaction pathways. Thereby, information about the evaluation of the mechanism of bonding could be explained by fitting sorption data of contact time experiments to the linear forms of pseudo-first-order, pseudo-second-order, intraparticle diffusion, pore diffusion ,and Elovich kinetics models [36] as shown in Table.2.

3.3.2.1. Pseudo-first-order model

The pseudo-first-order (PFO) model is also founded on the supposition that rate of adsorption is proportional to the sites available for adsorption and usually suitable for physisorbed species. The kinetics equation for pseudo-first order in linear form is

$$\log\left(q_e - q_t\right) = \log q_e - \frac{k_1}{2.303} t \qquad \text{(Eq. 12)}$$

The pseudo-first-order (PFO) kinetic model (Fig. 2A) for FUC-HG showed a significant difference between estimated (q_e,cal) and experimental (q_e,exp) adsorption capabilities, as well as a low regression coefficient (R^2 = 0.4099). Cd^{2+} adsorption does not follow a straightforward physisorption-controlled mechanism controlled by surface film diffusion, as evidenced by the divergence from linearity and the non-uniform distribution of residuals. Rather, first-order kinetics is insufficient to explain the quick initial uptake shown in experiments.

The chemical structure of FUC-HG, which has a high density of active sulfated ($–SO_3^-$) and hydroxyl (–OH) functional groups capable of significant ion-exchange and coordination interactions with Cd^{2+} ions, is compatible with the PFO model's restricted applicability. Adsorption occurs by a mixture of surface binding, intraparticle diffusion, and chemisorption since the hydrogel matrix is porous and extremely swellable. Therefore, the poor agreement with the PFO model indicates that strong site-specific chemical interactions rather than weak physical adsorption dominate Cd^{2+} removal by FUC-HG, supporting the appropriateness of higher-order kinetic models to describe the system.

3.3.2.2. Pseudo-second-order model

Heavy metal ions adsorption onto FUC-HG showed a pseudo-second-order (PSO) model and an excellent fit as indicated by the correlation coefficient (R^2 = 0.9876) in Fig. 2B. The model's linear form is written like this

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \qquad \text{(Eq. 13)}$$

In this equation, q_t is the amount of metal ions grabbed at time t in mg/g, q_e is the amount at the end in mg/g, and k_2 is the rate constant in g/mg/min.

According to the pseudo-second-order (PSO) kinetic model, chemisorption involving electron sharing or exchange between the adsorbent and adsorbate controls the rate-limiting step of adsorption. Cd^{2+} adsorption onto FUC-HG proceeds primarily through a chemically regulated mechanism rather than simple physical adsorption, as demonstrated by the excellent agreement between the experimental data and the PSO model. This robust model fit further demonstrates that particular interactions at the active binding sites, rather than just diffusion or external mass transfer, control the adsorption kinetics. The functional chemistry of the fucoidan hydrogel is intimately related to the applicability of the PSO model from a mechanistic standpoint. Sulfated ($–SO_3^-$) and hydroxyl (–OH) groups abound in the FUC-HG matrix, offering numerous coordination sites that can bind Cd^{2+} ions via ion exchange and complexation processes. In the three-dimensional hydrogel network, these functional groups serve as chemically active anchors that facilitate quick initial absorption and stabilisation once equilibrium is reached. Therefore, the dominance of PSO kinetics demonstrates the efficacy of FUC-HG as a strong and chemically active bioadsorbent for wastewater purification and reveals that

Cd^{2+} removal by FUC-HG is controlled by strong, site-specific chemical interactions.

3.3.2.3. Intraparticle diffusion model

The intraparticle diffusion model was also employed for a deeper comprehension of the rate-limiting step in Cd^{2+} adsorption onto the FUC-HG. To find out which step limited how quickly heavy metal ions stick to FUC-HG, we used the intraparticle diffusion model first suggested by Weber and Morris:

$$q_t = k_{idm} t^{1/2} + C \qquad \text{(Eq. 14)}$$

In this formula, kidm (mg g^{-1} $min^{-1/2}$) is the rate constant for diffusion into the particle, and C (mg g^{-1}) is the intercept of the line, which tells us about the boundary layer effect around the particle.

Cd^{2+} adsorption onto FUC-HG occurs via a multistep mechanism, as seen by the plot of q_t versus $t^{1/2}$ (Fig. 2C), which shows a quick initial absorption followed by a progressive plateau. Surface adsorption and boundary layer diffusion, where numerous and easily accessible sulfated binding sites on the hydrogel surface facilitate quick Cd^{2+} uptake, are responsible for the initial stage's rapid adsorption rate. A slower phase controlled by intraparticle diffusion ensues, during which Cd^{2+} ions move into the hydrogel network's interior pores and swelling domains.

The diffusion plot's nonlinearity and the lack of a straight line through the origin show that exterior film diffusion also plays a major role in the adsorption process and that intraparticle diffusion is not the only rate-limiting phase. When equilibrium is reached, the majority of active sites are occupied and additional Cd^{2+} transport into the hydrogel matrix is restricted. This is reflected in the observed plateau. Overall, these findings highlight the efficacy of the porous, sulfated hydrogel structure for effective heavy-metal removal from aqueous systems by showing that Cd^{2+} adsorption by FUC-HG is regulated by a combined mechanism involving surface adsorption, pore diffusion, and boundary layer resistance.

3.3.2.4. Pore diffusion

Bangham's equation helps us see how much pore diffusion affects how heavy metal ions stick to FUC-HG. The equation looks like this

$$q_t = \ln k_B + \alpha_B \ln t \qquad \text{(Eq. 15)}$$

Where k_B (m mg^{-1}) denoted the Bangham Constant whose value described the sorption maxima, α_B, is constant for pore diffusion whose value should be less than unity, (q_t/q =1)is the film diffusion constant (min^{-1}).

The Weber–Morris pore diffusion model was used to analyse the adsorption data in order to examine the role of intraparticle transport in Cd^{2+} uptake [37]. The plot of ln q_t versus ln t revealed a roughly linear connection (R^2 = 0.8628), suggesting that pore diffusion is important for the adsorption process in Fig. 2D. Nevertheless, the line did not cross the origin, indicating that surface adsorption and boundary layer resistance both affect the overall kinetics, indicating that adsorption is not only controlled by pore diffusion.

The nonzero intercept measures the initial resistance at the hydrogel solution interface, whereas the slope of the Weber–Morris plot indicates the rate of ion transport into the hydrogel pores. These findings lend credence to a progressive adsorption mechanism in which Cd^{2+} ions quickly attach to accessible surface sites, diffuse through the polymer's boundary layer, and then delve further into the internal pore network. The great efficacy of FUC-HG in sequestering Cd^{2+} ions from aqueous solutions can be explained by this layered adsorption behaviour, which is typical of biopolymer-based hydrogels and combines surface ion exchange with intraparticle movement.

3.3.2.5. Elovich model

Fig. 2E. shows how well the Elovich kinetic model describes metal-ion adsorption on FUC-HG. This model is ideal when adsorption occurs on surfaces with varying binding energies and is primarily controlled by chemical attachment. The Elovich model is given by

$$q_t = \frac{1}{\beta}\ln(\alpha\beta) + \frac{1}{\beta}\ln t \qquad \text{(Eq. 16)}$$

α (mg g^{-1} min^{-1}) is the initial adsorption speed, while β (g mg^{-1}) relates to how fully the surface can fill and how much energy is needed for the adsorption to occur. The

kinetics of Cd^{2+} adsorption on FUC-HG were assessed using the Elovich model, which produced a linear t/q_t vs lnt plot with a significant correlation coefficient (R^2=0.8887). This suggests that the hydrogel's surface is heterogeneous and contains active binding sites with different energies. An examination of the Elovich characteristics reveals a low desorption constant (β), indicating that ions remain firmly fixed by chemical interactions after adsorption, and a high initial adsorption rate (α), indicating the quantity of easily accessible sites for quick Cd^{2+} uptake.

The Elovich model's better fit than the pore diffusion model emphasises that surface reactions and chemisorption, rather than intraparticle diffusion, are the main factors influencing Cd^{2+} uptake. Strong ion-exchange and coordination contacts are facilitated by functional groups within FUC-HG, especially hydroxyl (–OH) and sulfated ($–SO_3^-$) moieties, which help remove Cd^{2+} ions effectively and selectively. These findings show that FUC-HG is an efficient bioadsorbent for wastewater treatment due to its highly reactive and diverse surface.

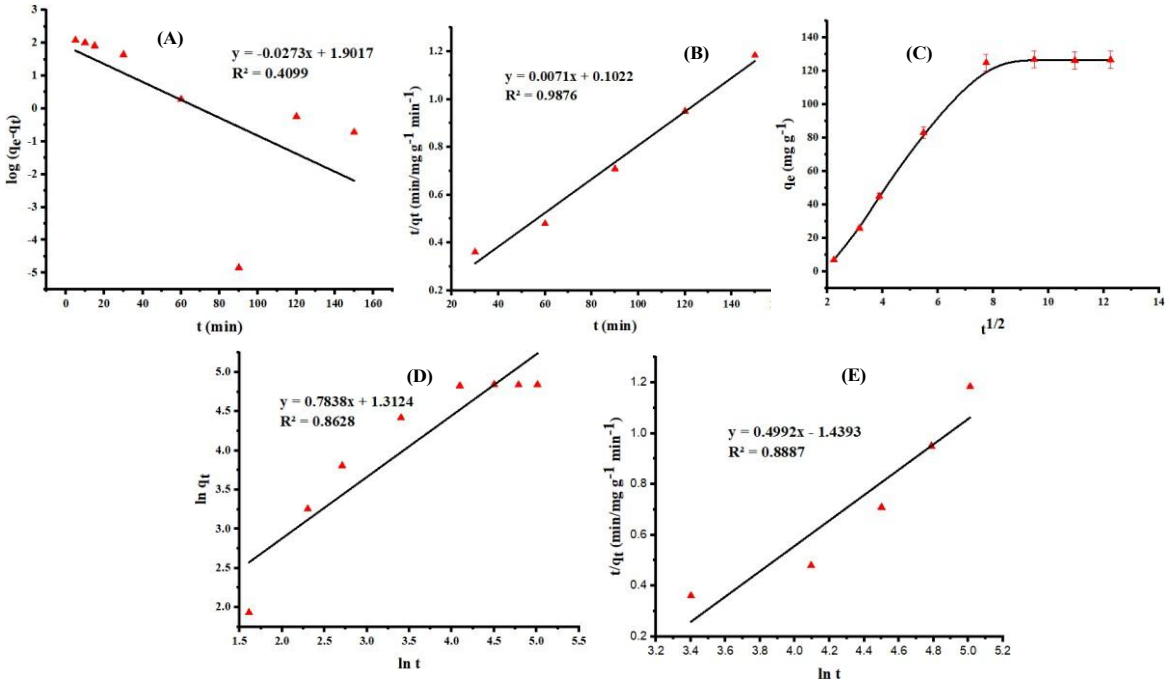

Fig. 2. Kinetic Models for adsorption of Cd^{+2} onto FUC-HG. **(**A) Pseudo-First-Order kinetics, (B) Pseudo-Second-Order Kinetics, (C**)** Intraparticle Diffusion model, (D) Pore diffusion, and (E) Elovich Model

Table. 2. The kinetics parameters of linear forms of pseudo-first-order, pseudo-second-order, intraparticle diffusion, pore diffusion, and Elovich models for the Cd(II) uptake by FUC-HG.

Experimental		q_e (mgg^{-1})	126.9143
Pseudo-first-order		q_e (mg g^{-1})	6.697203
		k_1 (g mg^{-1} min^{-1})	0.062871
		R^2	0.4099
Pseudo-second-order		q_e (mg g^{-1})	210.53
		k_2 (g mg^{-1} min^{-1})	2.20891E-05
		R^2	0.9876
Intra-particle diffusion model	Stage I	K_{id} (mg g^{-1} min$^{-0.5}$)	21.74002
		I (mg g^{-1})	-40.6063
		R^2	0.9958
	Stage II	K_{id} (mg g^{-1} min$^{-0.5}$)	0.320613
		I (mg g^{-1})	123.0066
		R^2	0.5229
Pore Diffusion		K_β	20.5288
		α_β	0.783823
		R^2	0.8628
Elovich isotherm		α (mg (g min)$^{-1}$ gmg^{-1}	0.02794
		β (mg (g min)$^{-1}$ gmg^{-1}	2.003092
		R^2	0.8887

3.4. Effect of Temperature on Cd^{+2} Uptake Beyond Thermodynamics

The experimental temperature-dependent performance gave additional mechanistic knowledge, even though thermodynamic parameters shed light on spontaneity and energetic nature. Raising the temperature enhanced intramolecular mobility in the hydrogel matrix, lowering diffusion resistance and increasing the accessibility of sulphate groups [38]. Wider diffusion channels were produced by the increased swelling seen at higher temperatures, which improved the penetration of hydrated Cd^{2+} ions. However, very high temperatures caused slight reductions in capacity, which could be explained by a partial rupture of the three-dimensional network of the hydrogel. The idea that the fucoidan hydrogel functions through a dual mechanism involving both surface binding and interior matrix diffusion is supported by this temperature-governed swelling diffusion connection.

3.4.1. Sorption Thermodynamics

For better understanding of the feasibility and the nature of the adsorption phenomenon, the thermodynamic parameters were determined by the temperature dependence of metal ion uptake on the FUC-HG in Table. 3. The values of these factors were calculated by using these equations [39].

The ΔG° is associated with the equilibrium constant (K_C) and was calculated using the given below Eq. 17. The $\Delta H°$ and ΔS° were calculated using Vant's Hoff isotherm (Eq. 19).

$$\Delta G^{o} = -RT \ln K_c \quad \text{(Eq. 17)}$$

$$K_c = \frac{q_e}{C_e} \quad \text{(Eq. 18)}$$

$$\ln K_c = \frac{\Delta S^{o}}{R} - \frac{\Delta H^{o}}{RT} \quad \text{(Eq. 19)}$$

A spontaneous and endothermic process is shown by the thermodynamic characteristics of Cd^{2+} adsorption onto FUC-HG. While the positive ΔH° indicates the endothermic nature, the negative ΔG° values validate the spontaneity of adsorption, indicating that higher temperatures improve Cd^{2+} transport into the hydrogel matrix because of greater

molecular mobility and stronger contacts with active sites. The displacement of solvated water molecules during ion binding is probably the cause of the positive ΔS°, which shows an increase in randomness at the solid solution interface. These findings support the kinetic analysis and demonstrate the dominance of chemisorption. Overall, the thermodynamic analysis shows that FUC-HG is a stable and effective bioadsorbent for the removal of Cd^{2+} that may function well in a variety of environmental settings.

Table. 3. Thermodynamic parameters of sorption of Cd(II) onto FUC-HG.

Thermodynamic parameter	$\Delta S°$ (J mol^{-1} K^{-1})	98.6
	$\Delta H°$ (kJ mol^{-1})	24.8
	$\Delta G°$ (kJ mol^{-1})	−4.58

3.5. Influence of Ionic Strength and Competing Ions

The uptake of Cd^{2+} was greatly impacted by the presence of background electrolytes, especially at higher ionic strengths where charge shielding decreased the effective electrostatic interaction between Cd^{2+} and the negatively charged sulphate groups. By directly competing for sulphate binding sites, divalent ions like Ca^{2+} or Mg^{2+} imposed more interference than monovalent cations like Na^{+}. However, the hydrogel maintained a significant amount of its adsorption capability even at high ionic strength, indicating the specificity of the sulphate cadmium interaction. This competitive performance suggests that the fucoidan hydrogel may perform well in actual wastewater matrices with mixed ions [40].

3.6. Proposed Adsorption Mechanism at the Molecular Level

Metal ligand chelation, intraparticle diffusion, and electrostatic attraction work in concert to control the adsorption of Cd^{2+} onto the FUC-HG. As the main binding sites, the sulphate groups ($–SO_3^–$) create persistent ionic or coordinate contacts with Cd^{2+} ions. In the hydrogel network, secondary contributions come from hydroxyl groups that offer extra coordination sites, forming bidentate or multidentate complexes [41]. Cd^{2+} is transported through interconnected holes in the swelling hydrogel matrix, where interactions with internal sulphate domains improve overall uptake. The strong affinity shown in experiments is explained by this hierarchical binding design, which also verifies that fucoidan functions as a naturally occurring ligand for metal ions because of its high degree of sulfation.

3.7. Structural Stability and Integrity of the Hydrogel During Adsorption

The hydrogel showed its resilience in aqueous environments by retaining its structural cohesiveness without appreciable fragmentation or disintegration across several adsorption cycles. Even when competing Cd^{2+} ions were present, the strong ionic crosslinking between fucoidan chains and Ca^{2+} maintained structural robustness. Rather than physical degradation, partial desorption and progressive occupation of deep matrix sites were found to be the main causes of minor reductions in adsorption capacity with repeated usage. The hydrogel's capability for scalable wastewater remediation was reinforced by the retained morphology and little loss of functional groups, which demonstrated the hydrogel's capacity to function consistently in repeated treatment cycles.

3.8. Comparative Performance Perspective with Natural and Synthetic Adsorbents

Even though FUC-HG was the study's main focus, its performance value becomes more apparent when compared to other adsorbents. Because of its high sulphate content, which offers stronger electrostatic and coordination interactions, the FUC-HG showed greater attraction towards Cd^{2+} when compared to traditional biopolymers like chitosan, alginate, or pectin. Fucoidan is a more sustainable substitute for synthetic polymeric resins, which are effective but can require complicated manufacture and raise

environmental issues. FUC-HG is a sophisticated bioadsorbent that may satisfy the needs of modern water treatment technologies because of its high Cd^{2+} absorption capacity, selectivity in the presence of competing ions, ease of regeneration, and biodegradability.

4. Conclusion

This work shows that FUC-HG, which is made from Undaria pinnatifida, is a selective and efficient bioadsorbent for removing Cd^{2+} from aqueous solutions. While kinetic analysis matched the pseudo-second-order model (R^2 = 0.9876), confirming that chemisorption is the major rate-limiting process, equilibrium adsorption data were best represented by the Freundlich isotherm (R^2 = 0.9441), indicating multilayer adsorption on a heterogeneous surface. The adsorption process is spontaneous, endothermic, and entropy-driven ($\Delta G° < 0$, $\Delta H° > 0$, $\Delta S° > 0$), according to thermodynamic analysis, which is in line with the chemisorption mechanism seen in comparable polysaccharide-based hydrogels documented in the literature. The hydrogel's promise for sustainable water treatment was highlighted by its outstanding stability and reusability, as well as its ability to remove Cd^{2+} efficiently across several cycles. FUC-HG is a potential material for heavy metal cleanup due to its high adsorption capacity (~50 mg/g), selective ion binding, and chemical resistance. In keeping with other published research on polysaccharide-based bioadsorbents, our findings indicate possible relevance in industrial wastewater treatment, including metal-contaminated effluents from electroplating or mining activities, even if the current results are at the laboratory scale. All things considered, FUC-HG is an economical, eco-friendly, and effective bioadsorbent for the targeted elimination of hazardous heavy metal ions, supporting the mounting evidence that sulfated polysaccharide hydrogels are workable options for water purification.

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