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Bioadsorbent (Rice Grains) Efficiency in Mercury II Removal from Aqueous Solutions: Adsorption Kinetics, Isotherm and Thermodynamics

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ABSTRACT

Heavy metals are major pollutants in marine, soil, industrial, and even treated wastewater. These metals are transported by flowing waters and polluted water sources downstream of the industrial site. Mercury is a highly toxically heavy metal. Mercury, on the other hand, is a highly toxically heavy metal. Mercury spillage is hazardous for it destroys the tissue, lungs, brain, and can affect the nervous systems and kidneys, causing some diseases. Therefore, removing Hg (II) from drinking water, aqueous solutions is essential in wastewater treatment and hydrometallurgical. A diverse process has been suggested to eliminate Hg (II) ions from wastewater. The adsorption method is used as a low-cost, efficient, and effective technique for removing toxically heavy metals from wastewater. Researchers have turned to inexpensive adsorbents such as vegetable wastes. This study aimed to remove Hg (II) ions from wastewater by using ground rice grains as adsorbents. The suitability of different isotherm and kinetic models for the adsorption process was researched. It was determined that the Langmuir isotherm best describes the adsorption equilibrium process, and the pseudo-second-order kinetic model is the most suitable model for adsorption. As a result of the analysis of thermodynamic parameters, it was concluded that the adsorption mechanism proceeds spontaneously and has an endothermic character. The data obtained show that rice grains can be considered a cheap, practical, and effective adsorbent for Hg (II) adsorption from wastewater.

Keywords:
Adsorption; Contamination; Isotherm; Kinetics; Mercury II; Thermodynamics

INTRODUCTION

The rapid increase in the world population, the excessive increase in industrialization, but the lack of environmental awareness in parallel with this rapidly consumes usable water resources. In addition to the advantages arising from the rapid rise in automation, it poses significant threats to the environment and living things due to the wastes produced. Harmful wastes produced by various industrial establishments can be divided into two categories as organic and inorganic wastes. Of these, organic wastes may be more unstable in the environment than inorganic wastes. However, inorganic wastes can remain undegraded for a long time, especially in aqueous environments, and can cause accumulations. When it comes to inorganic wastes, heavy metals usually come to mind [1]. Various definitions can be made for heavy metals. These metals are called "heavy metals" because their density is more than five g.cm³ in terms of their physical properties. More than 60 heavy metals can be identified in this way, including iron, copper, lead, zinc, nickel, cobalt, mercury, and chromium [2]. Heavy metals are among the pollutants that need to be removed due to their toxic and carcinogenic effects on human and aquatic environments through many sources. These dangerous pollutants are formed because of industrial, agricultural, waste disposal, and military activities. Industrial wastewater is the leading source of heavy metal pollution [3]. Some heavy metal salts are easily mixed in aqueous environments due to their excellent solubility in water. Since most of them dissolve in water as colorless, it is impossible to detect water contaminated with heavy metals easily. Heavy metal ions mixed with aqueous environments from various industrial establishments form precipitates as slightly soluble salts, especially in the sediments of seas, rivers, and lakes. Thus, when the heavy metal dissolved in the water decreases, the deposits release heavy metal ions into the water. This state shows how dangerous heavy metal pollution can be. In this way, living creatures living in waters polluted with heavy metal ions take heavy metals into their bodies. Thus, heavy metals in aquatic environments pass from the most minor living thing to the body of other living things with an increasing concentration through the food chain and threaten [4].

Some conventional techniques of dealing with aqueous contamination include adsorption, coagulation, precipitation, membrane separation, reduction, photocatalysis, ion exchange, and so forth [5, 6]. Amongst these techniques, the adsorption process is easy, low-cost to and an effective way to handle water pollution [7]. Therefore, the quality and quantity necessities of adsorbents are rising. Recently, there are many kinds of commercially present adsorbents for different practices. The improvement of environmentally more efficient, cheap, and friendly adsorbents is a very active investigative issue. Therefore, alternative low-cost adsorbents such as chitin [8], coffee [9], tea waste [10], rice husk [11], orange peel [12], bark [13], and coir pith [14] have been studied.

A widely sourced and low-cost adsorbent with high adsorption capacity should be used to remove mercury ions, a toxic heavy metal, from wastewater. Grown rice is the second most grown cereal plant globally and significant fundamental food for more than half of the world's population [15]. Rice contains some unique ingredients that have proven benefits for human health. Rice grains contain phenolic compounds with antioxidant activity. The most widespread shapes of phenolic compounds in rice are hydroxybenzoic and hydroxycinnamic acids. Other compounds defined contain protocatechuic acid, sinapic, and p-hydroxybenzoic acid, which are benzoic acids. Additionally, the two primary groups of compounds influential, aldehyde analogs such as vanillin are also called phenolics. [16].

This research aims to specify the optimum parameters for the maximum adsorption of Hg (II) ions from aqueous solutions using rice grains, which are widely used and contain different compounds in their structure as adsorbents. In addition, using the obtained data, adsorption kinetics, isotherm models, and thermodynamic properties were evaluated.

MATERIAL AND METHODS

Chemicals and instruments

The rice grains used were purchased from local markets. The $HgCl_2$ salt, 1,5-diphenylcarbazide, Acetic acid (CH_3COOH), and Sodium hydroxide (NaOH) were obtained from Merck and were of analytical grade. A stock solution of mercury chloride salt was prepared (500 mg/L) and diluted to desired concentrations.

WiseStir multiple mechanical stirrer heater, NUVE FN 400 oven, Thermo Scientific ultrapure water device, 620 Lab pH Meter, Optizen POP UV spectrophotometer were used in the experiments.

Adsorption Experiments

Rice obtained from local markets was ground into particles of approximately 100-150 mesh and dried in an oven at 50°C for 12 hours to remove moisture. Dried rice grains were stored in a desiccator for use in experiments without any further modification. Mercury ions adsorption studies were carried out with bioadsorbent (rice grains) in Hg (II) solution (10 mL). The pH of the Hg (II) solution is adjusted between 3 and 10 using diluted 0.1 M CH_3COOH or 0.1 M NaOH solutions. A known mass of bioadsorbent was then added to the mercury solution and shaked at 500 rpm and room temperature. After the adsorption process, the bioadsorbent was removed from mercury solutions by filtration. The concentration of Hg (II) in the solutions was determined at 532 nm with a UV spectrophotometer using 1,5-diphenylcarbazide [17]. The effects of process variables like contact time, temperature, pH, and initial concentration on the mercury removal efficiency were investigated.

Equations for determining the amount of Hg (II) adsorbed using bioadsorbent are given below:

$$q_t = \frac{(C_0 - C_t)}{m} xV \quad (1)$$

$$q_e = \frac{(C_0 - C_e)}{m} xV \quad (2)$$

$$\%R = \frac{(C_0 - C_t)}{C_0} x100 \quad (3)$$

Where, q_e and q_t are the adsorption capacity (mg/g) at equilibrium and t, respectively; C_o, C_e and C_t, initial concentration at time t, mercury equilibrium concentration (mg/L) and liquid phase concentration, respectively; the volume (L); m is the amount of adsorbent (g); and R is removed yield (%) [18].

RESULTS AND DISCUSSION

Infrared Spectrum of Bioadsorbent

FT-IR spectra of the bioadsorbent before and after adsorption were recorded between 4000 and 400 cm^{-1} using Bio-Rad-Win-IR spectrophotometer. When the FT-IR graphs before and after Hg (II) adsorption are examined (Fig. 1); stretching vibration of hydroxyl (O-H) and N-H bonds around 3600 cm^{-1}; at 2970 cm^{-1} the asymmetric stretching of the –CH– groups; asymmetrical carboxy-

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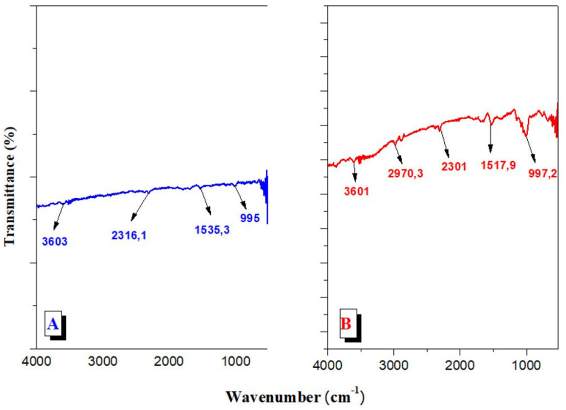

Figure 1. FTIR plots of bioadsorbent (a) after and (b) before Mercury II adsorption

late (-COO-) stretching vibration at 1517 cm^{-1}; -C–O stretching vibration of alcohol, phenol and carboxylic groups is observed at 997 cm^{-1}. It was observed that the peaks of the functional groups of the bioadsorbent determined before the adsorption process changed because of Hg (II) adsorption. It was observed that some of the previously determined peaks disappeared after Hg (II) adsorption, and there was a shift in frequency values at some peaks. This proves that physical or chemical bonds are formed between the surface functional groups of the bioadsorbent and Hg ions and that adsorption takes place.

pH Effect

The effect of initial pH on Hg (II) removal efficiency was investigated in the pH range of 4-10. Maximum adsorption (84.58%) was obtained at pH=6 (Fig. 2). Depending on the pH range, three main types of $HgCl_2$, HgO, and Hg(OH)Cl can be found in the solution. Under acidic conditions, $HgCl_2$ is present, and protons compete with mercury ions to occupy active sites. Thus, the uptake of $HgCl_2$, mercury ions is reduced. In the pH (4-6) range, Hg (OH)Cl is formed, a species that increases mercury uptake. Under primary conditions, red mercuric oxide precipitates with the most thermodynamically steady kinds obtained at high pH [19]. In a study on the adsorption of mercury on granular activated carbon in aqueous solutions containing nitrates and chlorides, the optimum pH value was found to be 5 [20].

Effect of Adsorbent Amount

In the experiments carried out to examine the effect of the adsorbent dose on the adsorption of mercury ions; Varying amounts of adsorbent in the range of 25-150 mg were used. According to the results obtained, maximum adsorbance was obtained when 100 mg of adsorbent was used and this amount was also used in subsequent studies. The results are shown in Figure 3.

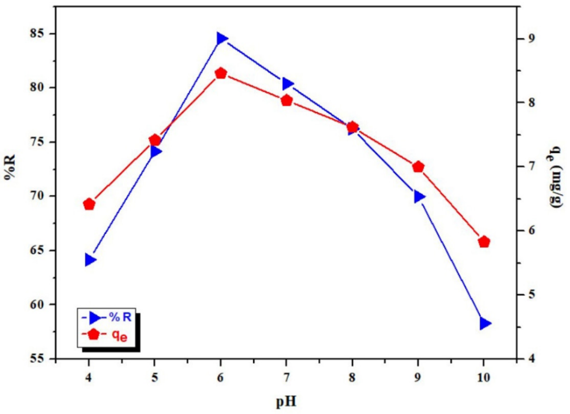

Figure 2. The impact of pH on Mercury II adsorption (25°C, 60 min, 50 mg/L, pH: 4-10).

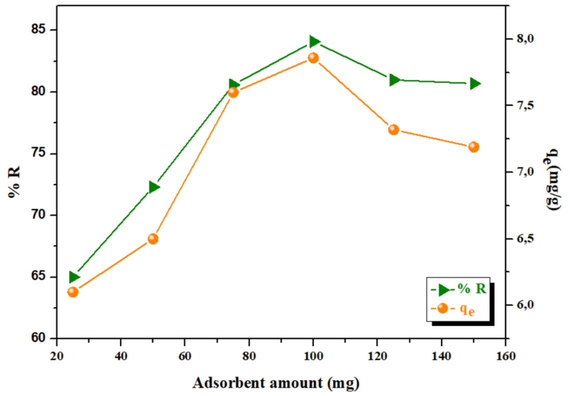

Figure 3. The impact of adsorbent amount on Mercury II adsorption (25°C, pH: 6, adsorbent: 25-150 mg, 50 mg/L).

Effect of Contact Time and Kinetic Studies

One of the parameters effecting adsorption is contact time. As a result of adsorption of 100 mg bioadsorbent and 50 mg/L $HgCl_2$ solutions at pH 6.0 at 25°C and mixing at 500 rpm for 5-180 minutes, the maximum contact time was found to be 30 minutes (Fig. 4). This time was also used in subsequent parameter studies.

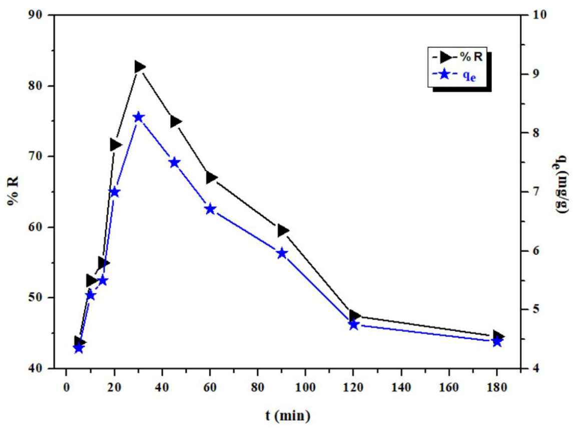

Figure 4. The impact of contact time on Mercury II adsorption (pH: 6, 25°C, 100 mg adsorbent, 50 mg/L).

The kinetics of Hg (II) adsorption were investigated with Pseudo first order (Fig. 5a) [21] and Pseudo second order (Fig. 5b) [22], and intra-particle diffusion (Fig. 5c) [23] models according to the following equations and the calculated parameters are shown in Table 1.

$$\ln(q_e - q_t) = \ln q_e - k_1.t \quad (4)$$

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

$$q_t = K_{id}.t^{1/2} + I \quad (6)$$

Where, q_t and q_e are the adsorption capacity (mg/g) at t and equilibrium, respectively. k₁ (1/min) and k₂ (g/mg.dk), the ratio constant of the pseudo-first-order and pseudo-second-order model, K_{id} particle inside diffusion rate regular (mg/g.min$^{1/2}$). The value of I gives an idea of the thickness of the boundary layer. That is to say, the greater the intersection degree, the greater the boundary layer effect.

Table 1. Kinetic parameters of Mercury II adsorption on bioadsorbent.

Models	Parameters	Hg (II)
Pseudo-first order	q_e (mg/g)	6.36
	k_1 (1/min)	0.07
	R^2	0.8527
Pseudo-second order	q_e (mg/g)	8.19
	k_2 (g/mg.min)	0.02
	R^2	0.9935
Weber-Morris intra-particle diffusion	q_{exp}	8.27
	k (mg/gmin$^{1/2}$)	1.08
	I (mg/g)	1.8
	R^2	0.8937

When Table 1 is examined, the pseudo-second-order kinetic model appears to be the most appropriate for Hg (II) adsorption. This equality is used to explain the adsorption behaviors at lower concentrations. In the table, it is seen that

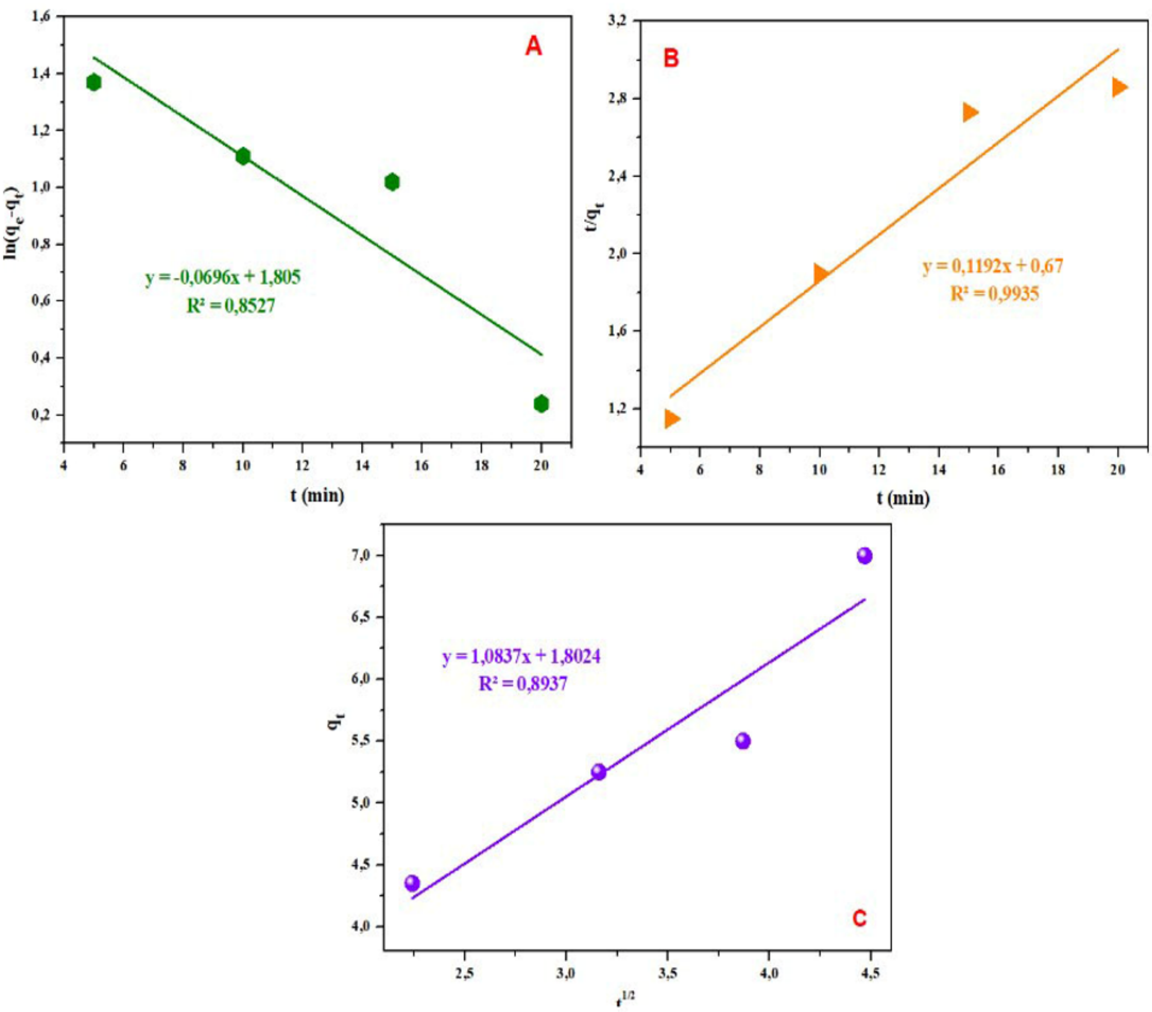

Figure 5. Adsorption kinetics of Mercury II adsorption on bioadsorbent according to (a) pseudo-first-order, (b) pseudo-second-order, (c) intraparticle diffusion kinetic models.

the correlation coefficient R^2 of the pseudo-second-order equation is close to 1 (0.9935), and the q_e value (8.19) calculated from the equation gives a result closer to the q_{exp} (8.27) value. The inadequacy of the pseudo-first-order rate equation to fit the kinetic data is due to the boundary layer that controls the initiation of the adsorption process. In a study in which toxic mercury was removed from petroleum oil using a molecularly imprinted polymer, it was stated that the adsorption process was suitable with the pseudo-second-order kinetic model [24].

Initial Concentration Effect and Adsorption Equilibrium Isotherms

The effect of the initial mercury concentration on the adsorption efficiency was investigated using $HgCl_2$ solutions with concentrations ranging from 10 to 250 mg/L. In the given concentration range at pH 6.0, mercury solutions were shaken with 100 mg bioadsorbent at 500 rpm for 30 minutes at 25°C. Optimum adsorption efficiency (85.8%) was obtained at 50 mg/L concentration.

Experimental data analyzes were performed in Langmuir, Dubinin-Radushkevich, Temkin, and Freundlich isotherm models to evaluate the adsorption equilibrium process [25, 26, 27, 28]. Isotherm graphs are given in Figure 6, and isotherm parameters are given in Table 2.

The compatibility of Langmuir, Dubinin-Radushkevich (D-R), Temkin, and Freundlich isotherm models to the mercury ion adsorption mechanism was investigated and it was determined that Langmuir isotherm was more compatible. The correlation coefficient calculated for the Langmuir isotherm was R^2= 0.9964, which was higher than the other isotherms. Accordingly, in the Langmuir equilibrium isotherm, the adsorption process is adsorption in which mercury ions bind to functional groups and binding sites homogeneously distributed on the adsorbent surface in a monolayer. At the

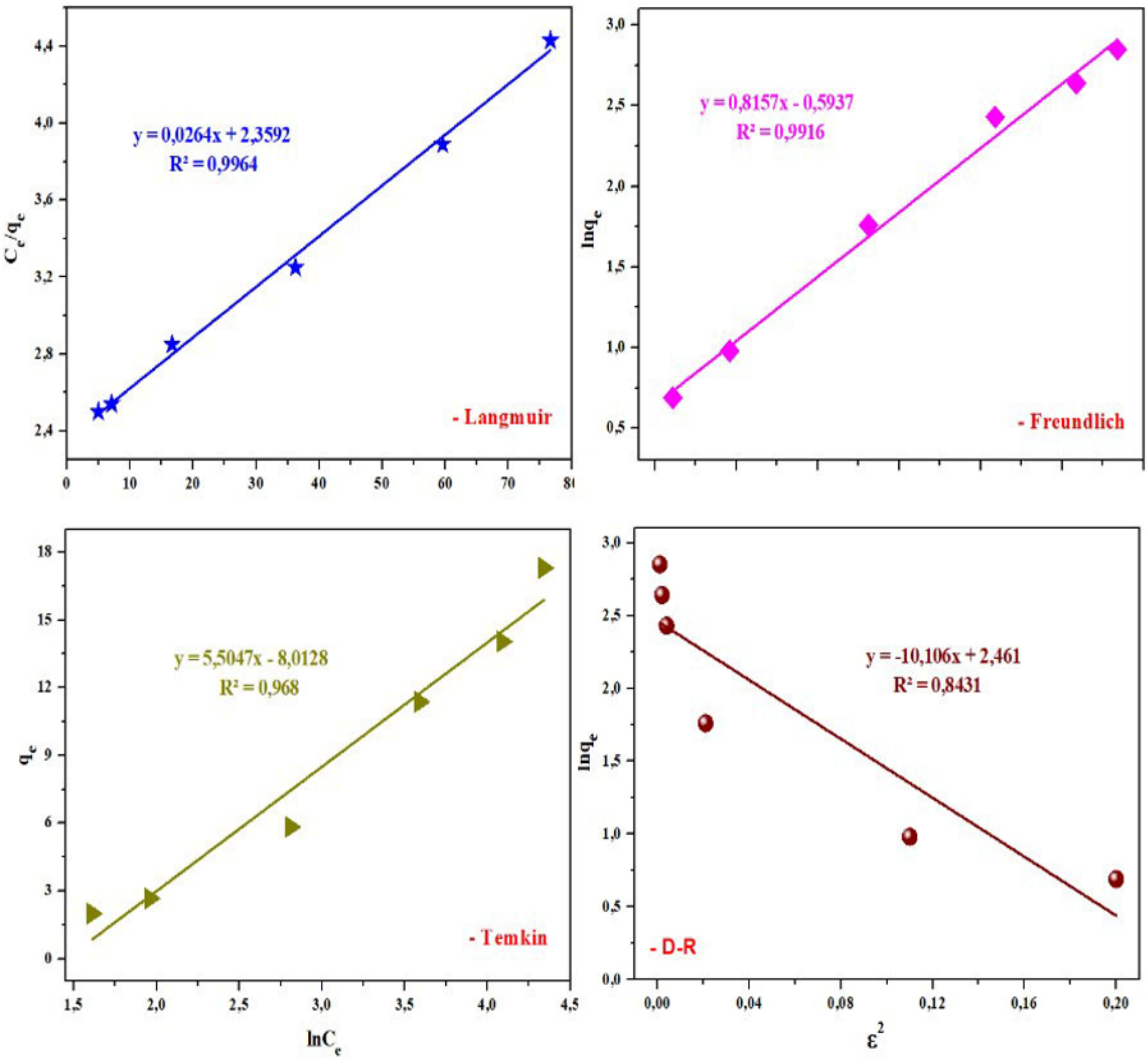

Figure 6. Isotherm graphics for the Mercury II adsorption process.

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Table 2. Isotherm parameters for adsorption process.

Models	Parameters	Hg (II)
Langmuir	q_{max} (mg/g)	37.88
	b (L/mg)	0.01
	R_L	0.2
	R^2	0.9964
Freundlich	Kf [(mg/g) (L/mg)$^{1/n}$)]	0.55
	n	1.23
	R^2	0.9916
Temkin	bT	0.23
	K_T (L/g)	5.5
	R^2	0.968
Dubinin-Radushke-vich	q_m(mg/g)	11.72
	β (1/mol².J²)	10.1
	E (J/mol)	0.22
	R2	0.8431

same time, in this adsorption, there is a certain number of active sites on the adsorbent surface and the molecules bind to these active ends. Accordingly, mercury ions are attached to the functional groups and binding sites on the surface as a single layer. The maximum monolayer adsorption coefficient (q_{max}) value was found to be 37.88 mg/g. Using the b value from the calculated Langmuir parameters, the R_L values (dimensionless separation factor) were found to be as 0.2. The fact that these values are in the range of $0<R_L<1$ indicates that the adsorption mechanism is suitable for mercury ions. In a study in which mercury adsorption was performed by rice husk ash, it was stated that Langmuir isotherm, one of the isotherm models, was more compatible with the adsorption system and the maximum adsorption capacity (q_{max}) was 9.32 mg/g [29].

Then value defined in the Freundlich isotherm model was calculated as 1.23. The calculated n value in the range of 1<n<10 means that the adsorption of mercury ions on the bioadsorbent is favored and positive.

The mean adsorption energy (E) value defined in the Dubinin-Radushkevich isotherm delivers an opinion concerning the reaction mechanism that is efficient in the adsorption process. Since the calculated E value is calculated to be 0.22 kJ/mol (E<8 kJ/mol), physical forces are efficient in adsorption. Due to physical adsorption, it can be mentioned that there are weak Van der Waals attraction forces between bioadsorbent and mercury ions.

Temperature Effect and Thermodynamic Studies

To examine the impact of ambient temperature on the adsorption of 50 mg/L solutions of mercury ions with 100 mg bioadsorbent; It was studied for 30 minutes at pH 6.0, at a stirring speed of 500 rpm, at temperatures ranging from 20-50°C. The optimum temperature for the adsorption process was found to be 50°C (Fig. 7). The rising in

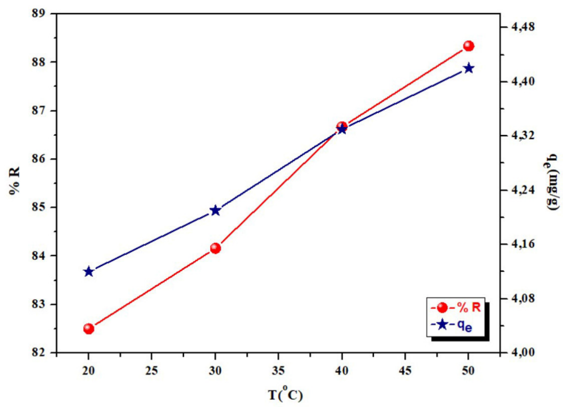

Figure 7. The effect of temperature on Mercury II adsorption (pH: 6, 30 min, 100 mg adsorbent, 50 mg/L).

adsorption yield with temperature indicates that the process is endothermic.

The thermodynamic parameters enthalpy change (ΔH°), free energy (ΔG°), and entropy change (ΔS°) are geted from the following equations [30].

$$\ln K_d = \frac{\Delta S^o}{R} - \frac{\Delta H^o}{RT} \quad (7)$$

$$\Delta G^o = \Delta H^o - T\Delta S^o = -RT \ln K_d \quad (8)$$

$$K_d = \frac{C_{ad}}{C_e} \quad (9)$$

Inequality, C_e is the equilibrium concentration of mercury in the solution; K_d is the equilibrium constant; C_{ad}, the concentration of mercury adsorbed to the adsorbents at equilibrium; R (8.314 (J/mol.K) is the gas constant; T (K) is temperature. ΔH° and ΔS° are obtained from the incline and intersection of the Van't Hoff plot of $\ln K_d$ for 1/T (Fig. 8), values are obtained from the slope and intersection of the graph, and these results are shown in Table 3.

The enthalpy of adsorption (ΔH°) value calculated from the incline of the line in the graph in Figure 8 was favorable for the adsorption of mercury ions. It showed that the adsorption had an endothermic nature. At the same time, the fact that the adsorption enthalpy values are lower than 40 kJ/

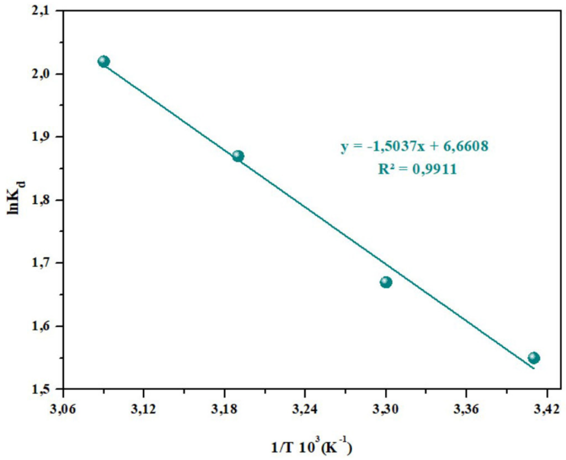

Figure 8. Change of ln K_d with 1/T in Mercury II adsorption.

Table 3. Thermodynamical parameters for Mercury II adsorption.

Thermodynamic parameters					
	T (K)	ΔH^0 (J/mol)	ΔS^0 (J/mol K)	$T\Delta S^0$ (kJ/mol)	ΔG^0 (kJ/mol)
Hg (II)	293	12.5	55.38	16.4	-3.77
	303			16.78	-4.21
	313			17.33	-4.87
	323			17.89	-5.42

mol shows that the adsorption processes can be explained by physical adsorption. The adsorption entropy (ΔS°) value calculated from the shift of the graph in Figure 8 was also found to be positive. This result shows that the adsorption occurs spontaneously and that the bioadsorbent has an affinity for mercury ions. The Gibbs free energy change (ΔG°) was calculated as an average of -4.57 kJ/mol. The increase in the negative values of ΔG° with rising temperature indicates that adsorption at high temperatures is even more convenient and applicable. At the same time, the negative value of Gibbs free energy (ΔG°) shows that adsorption consists spontaneously. In a study in which mercury adsorption was carried out on a carbon sorbent obtained from fruit peel; It was stated that the positive value of ΔH° indicates the endothermic adsorption of Hg ions and the positive value of ΔS° indicates the increasing randomness at the solid-solution interface during Hg (II) adsorption. It was also stated that the negative values of ΔG° and the decrease in ΔG° with temperature increase indicate the spontaneous nature of its adsorption [31].

CONCLUSION

The adsorption method is a very effective method among the various methods used for the removal of heavy metals in wastewater, nowadays, where the prevention of environmental pollution is becoming increasingly important. In the study, rice grains containing different compounds in their structure were used as adsorbents to remove toxic metal pollutants.

The suitability of adsorption with isotherm models, kinetic models, and thermodynamic expressions was investigated with the data obtained due to the experiments for the adsorption of mercury ions from aqueous solutions on bioadsorbent.

Pseudo-first-order kinetic model, pseudo-second-order kinetic model, and intraparticle diffusion models were evaluated for mercury adsorption processes. The most suitable kinetic model was determined to be the pseudo-second-order kinetic model. Freundlich, Langmuir, Dubinin-Radushkevich, and Temkin's isothermal models were examined for their suitability for adsorption processes. It was stated that the equilibrium data of adsorption isotherms were quite compatible for the Langmuir model in the concentration range studied, and the qmax value was calculated as 37.88 mg/g. As a result of the analysis of thermodynamic parameters, it was determined that the adsorption processes were spontaneous and endothermic. Also, entropy with a positive value shows the affinity of mercury ions to the bioadsorbent.

According to the results, it is stated that rice grains used without any chemical treatment to remove mercury from aqueous solutions are low cost and effective adsorbents. It is thought that this bioadsorbent can be used as an alternative to adsorbents prepared with costly and complex processes, and this study will contribute to environmental studies.

CONFLICT OF INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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