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Adsorption study of Hg(II) from Aqueous Solution by using Refused Vetiver Grass Root

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Abstract

The need for reasonably priced adsorbents with the ability to adsorb Hg(II) ions from industrial waste waters and effluents is rising. Consequently, the plentiful lignocellulosic material that was rejected from aroma crop extraction plants and industries. As a byproduct, vetiver grass (VG) root is used to bind mercury ions from lab solutions that have been prepared with khus oil. According to the Langmuir model, VG has a significant potential capacity (87.5 mg/g) at optimized pH 5.0 for the removal of Hg(II) ions. The pseudo-second-order model for the kinetic study was found to best support the adsorption process. The adsorption mechanism is well explained based on supporting results obtained from adsorption isotherms and FTIR analysis. Surface complexation/ion exchange is involved in VG adsorbent. The adsorption experimental process was studied with the application of various influencing factors, i.e., Hg(II) ion concentration, contact time, pH effect, and adsorption isotherms.

Keywords : Hg(II), adsorption, Refused vetiver root grass, batch studies, mechanism

1. Introduction

Mercury (Hg(II)) is one of the most hazardous heavy metals due to its high persistence and ability to bioaccumulate, [1], having negative health consequences on organisms with prolonged exposure [2]. Anthropogenic activities like fuel consumption and artisanal gold mining cause high amounts of mercury ions to seep into soil and water bodies [3, 4]. Consequently, the highest allowable amounts of mercury (II) in drinking water have been determined by the World Health Organization (WHO) and the United States Environmental Protection Agency (USEPA) to be 0.002 mg L^{-1} and 0.001 mg L^{-1} , respectively [5, 6]. Because mercury ions are extremely toxic, it is necessary to cleanse contaminated water to get rid of them. The most popular and efficient

techniques for removing mercury (II) from contaminated or industrial effluents in order to adhere to legally permissible limits are membranes, biosorption, ion exchange, reverse osmosis, and chemical precipitation. These are just a few of the numerous processes used to purify the contaminated water [7]. But there are few drawbacks in these techniques, such as being more costly, the operation system is not easy, it requires more energy, and it produces a lot of sludge or creates a lot of byproducts even though they work effectively. [8]. Adsorption is one of the most promising methods due to its high efficiency, low cost, and simplicity of usage in wastewater treatment facilities. It is necessary to produce affordable, selective materials with high adsorption capacities for these reasons [2]. Consequently, the development of an appropriate treatment approach that is affordable, easy to use, and environmentally friendly is required [9]. Because it is the most cost-effective and efficient method when compared to other approaches, adsorption has certain drawbacks, including the need for chemical regeneration, the high cost of preparing the adsorbent (such as in the case of activated carbon), and the adsorbent's loss of adsorption capacity with each regeneration cycle [10]. It may be more affordable to manage waste materials rather than remove heavy metal ions from contaminated water. Natural resources, byproducts, rejected agricultural waste, or industrial waste may be suitable options. For the creation of high-potential adsorbents, cheap and abundant raw materials are essential. Adsorbent materials can generally be considered "low-cost adsorbent" if their production involves simpler and quicker processing, they are widely accessible, or they are leftover materials from other industries or agricultural wastes [11]. A wide range of adsorbents, such as sugar beet pulp, coal fly ash, coir pith, rice husk cellulose, castor tree leaf powder, etc., are derived from natural resources and can be utilized to treat harmful pollutants such heavy metals, dyes, and detergents as well as polluted water [12–16].

Here, vetiver, also called Khus-khus grass and Vetiver in its native Western and Northern India, is a perennial grass whose roots yield an essential oil that is frequently used in aromatherapy, cosmetics, and perfumes. By using either steam distillation or hydro distillation, vetiver root grass yields vetiver oil. There is a plentiful supply of discarded vetiver root grass near the distillation facilities where fragrance components are produced. If Hg(II) ions need to be extracted from the mercury-contaminated water, VG might be a suitable substitute. The

concentration of Hg(II) in the solution, the pH of the Hg(II) ion solution, and the total amount of time needed to attain equilibrium at maximum adsorption capacity are some of the affecting parameters that are applied to optimize and explain the adsorption process. Studied the adsorption isotherm and response rate using kinetic and isothermal models of adsorption. Kinetic models, adsorption isotherms, and FT-IR analysis define and validate the binding of Hg(II) ions to the surface of VG.

2. METHEDOLOGY

2.1 Preparation of synthetic solutions

Hg(II) chloride (1.354 g) was dissolved in deionized water (DIW) and diluted to a volume of 1 L to provide a stock solution of 1000 mg/L. India-based Merck provided. AR grade comprised all other reagents.

2.2 Preparation of Adsorbent

The industrial estate area of the aroma crops and material distillation factories in Makrandnagar, Kannauj, Uttar Pradesh, India, was the source of discarded vetiver grass root (VG). VG was washed with tap water thoroughly to remove dust and unwanted dirt. They were initially dried in an oven for 72 hours at 50 °C before being ground into 60–100 mesh sizes. Hexane was used in a solvent-soxhlet extraction process for six hours to first dewax the dried VG powder. In order to adsorb the Hg(II) ions as an adsorbate, dried and solvent-free VG powder was sieved and kept in a desiccator.

2.3. Experiment for the adsorptive removal of Hg (II)

Batch adsorption processes were used to carried out the adsorption potential of the VG for the adsorption of Hg(II) ions from the synthetic Hg(II) aqueous solution. For the optimizing adsorption process as higher adsorption capacity or higher percentage of removal. The ideal conditions i.e. pH, contact time, concentration of adsorbate and adsorbent, were observed for achieving maximum Hg(II) ions removal. The powdered VG (0.025 g) of adsorbent was added to Hg(II) ions solution (50 ml) by the concentration (12.5-100 ppm) in a Erlenmeyer flask(100 ml). In order to examine the influence of pH (2–6) was studied by Using NaOH(0.1 N) and HCl(0.1M) solution before the adsorbent was added in order to examine the impact of the pH. The VG (0.025g) was combined with the Hg(II) ions solution in an Erlenmeyer flask and shaken for two hours at 120 rpm using a electric shaker. After that, a Whatman-41 filter paper was used

to filter the suspension. The unadsorbed concentration of Hg(II) ions was analyzed by using the atomic adsorption spectrophotometer (AAS). Equations (1) and (2) were used to determine the equilibrium adsorption capacity (q_e) (mg/g), or the amount of Hg(II) adsorbed per unit mass of the adsorbent, and the percentage of Hg(II) removal (R).

$$q_e = \left(\frac{C_i - C_e}{m} \right) V \quad (1)$$

$$q_e (mg/g) = \left(\frac{C_i - C_e}{M} \right) V \quad (2)$$

where C_i (mg/L) is the initial concentration of Hg (II); C_e (mg/L) is the unadsorbed concentration of Hg (II); V (L) is the adsorbate volume; and M (g) is the weight of the adsorbent.

3. RESULTS AND DISCUSSION

3.1. Role of concentration of adsorbate, pH and contact time

The percentage of Hg (II) ion removal was measured by increasing Hg (II) concentrations from 12.5 to 100 mg/L while 0.025 g constant adsorbent doses of VG were swirled. The relationship between concentrations (12.5 to 100 mg/L) and adsorption capacity (mg/g), or the percentage of mercury ion removal, is shown in Figure 1(a). The percentage removal of Hg(II) drops from 99.6% to 76.4% at a constant adsorbent concentration (0.025 g) when the concentration of Hg(II) ions rises from 12.5 to 100 mg/g. The amount of Hg(II) that is adsorbed per unit mass of VG rises proportionately to the rise in the concentration of Hg(II) in the aqueous solution. The lowest amounts of Hg(II) adsorbed were determined to be 24.9 mg/g, with starting concentrations ranging from 12.5 to 100 mg/L, respectively.

Figure 1(b) illustrates the pH range in which the adsorption of Hg(II) onto VG was investigated. The pH of the adsorbate synthetic Hg(II) ions standard solution has a significant impact on the adsorption of Hg(II) ions. Adsorption efficiency rises quickly at first and subsequently decreases as the solution's pH rises. It was discovered that the largest percentage removal was from solutions with pH values between 2 and 5, or 99.6% for pH 3 and 99.2% for pH 4 and pH 5, respectively. The poor adsorption of Hg(II) ions at low solution was caused by the competitive coordination action of H^+ ions with COOH-donor atoms of -OH and O donor atoms of -OH on the surface of adsorbents.

Furthermore, Hg(II) adsorption may be difficult at lower pH levels because VG's functional groups may mostly persist in the protonated state. An experiment up to 180 minutes in length was conducted to find the equilibrium contact time. Based on the observation (as illustrated in Figure 1(c)), it was found that the removal effectiveness rises with increasing duration from 15 to 180 minutes, rising from 75.5 to 97.6% before stabilizing and reaching the stationary phase. Over a 180-minute period, the metal removal efficacy does not change due to saturated receptors on the surface of the VG adsorbent. Instead, by agitating 50 mL of standard Hg(II) aqueous solution containing 100 ppm of Hg(II) at pH 5, the ideal contact time for 0.025 g of VG is determined to be a minimum of 180 minutes (3 hours).

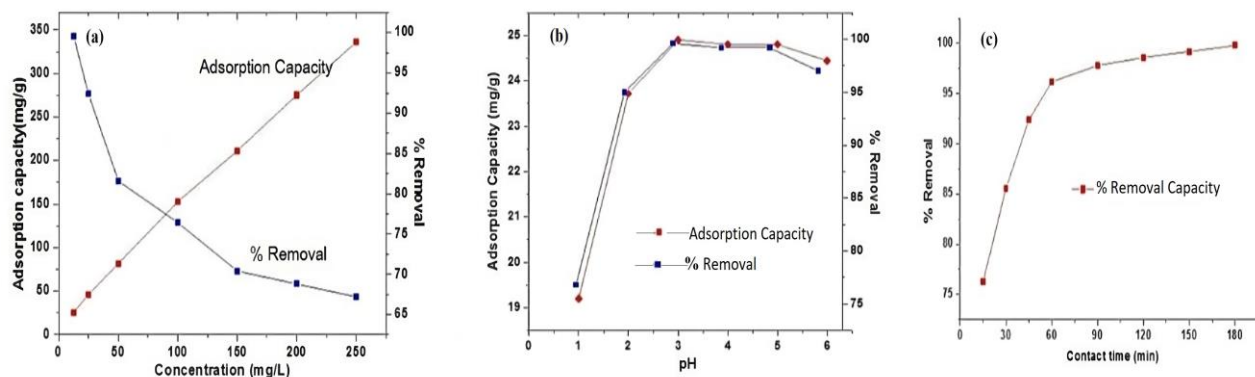


Figure 1. (a) Effect of adsorbate (mercury ions) concentration (b) Effect of pH and (c) Effect of contact time for the adsorption Hg(II) ions.

3.2 Elemental/ CHNS analysis

To understand an adsorbent's behavior during the adsorption process, it is vital to understand its physicochemical composition. Physisorption and chemisorption are two primary processes in adsorption, where pH, moisture content, solubility in water and HCl (0.25 M), and percentages of hydrogen (H), nitrogen (N), sulfur (S), and oxygen (O) concentrations are important factors. Table 1 reflects the percentage of the C, H, N, S, and O analyses in the VG.

Table 1. Physical and chemical Properties of the VG.

Parameters			Value		
pH			6.7		
Moisture content (%)			13.6		
Solubility in water (%)			0.0		
Solubility in 0.25 M HCl (%)			0.0		
Ultimate analysis/mass %					
Element analysis	C%	H%	N%	S%	O
VG	41.90	5.94	0.88	-	51.28

3.4 Determination of Functional groups

The presence of significant peaks at $3600.05\text{--}3100.01\text{ cm}^{-1}$ demonstrated to lignin as shown in Figure 2, which is in the raw VG, Fourier-transform infrared spectroscopy (FTIR) spectrum. O-H peak is due to the stretching intermolecular hydrogen bonds. The presence of bending vibration of -OH is shown by the peaks at 1554.99 and 1638.9 cm^{-1} . The C-H band stretching peaks 1465 and 1412 cm^{-1} are defined the presence of cellulose's. Bending vibrations of -C-O are responsible for the peak at 1250 cm^{-1} . Following the adsorption of Hg(II) ions, the FTIR spectra of vetiver grass exhibit a significant shift in the peak for O-H, which was previously located around 3599.01 and 3426.05 cm^{-1} , but is now shifted to 3546.80 and 3432 cm^{-1} , respectively. The peak of -OH at 1639.60 cm^{-1} was slightly moved to 1642 cm^{-1} due to the presence of mercury ions. Because of the Hg(II) ions, the peaks at 1556 cm^{-1} and the peak of the -C-O bending vibrations at 1250 cm^{-1} are moved to 1549.80 cm^{-1} and 1304 cm^{-1} , respectively.

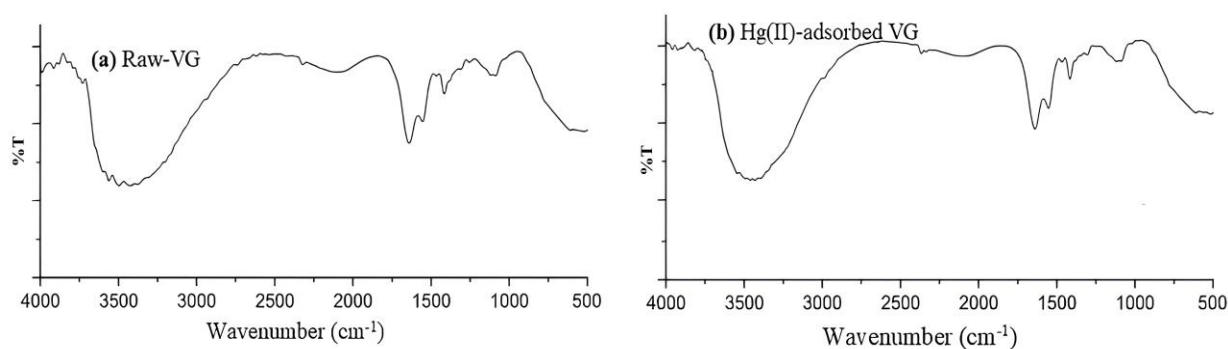


Figure 2. Fourier-transform infrared spectra: (a) raw VG (b) Hg(II) adsorbed-VG

3.5 Study of adsorption isotherm: Langmuir and Freundlich's isotherms

The relationship between Hg(II) ions and VG surface active sites was investigated using Langmuir and Freundlich's isotherms. The Langmuir adsorption model is well recognized to be applicable to monolayers, or materials with comparable adsorption potential at active sites. In Eq. 2, the Langmuir isotherm is expressed as follows:

$$\frac{C_e}{q_e} = \frac{1}{bK_L} + \frac{C_e}{b} \quad (2)$$

Maximum quantity of Hg(II) ions per unit weight of VG is denoted by $q_{\max}$, while q_e represents equilibrium. At equilibrium, C_e represents the unadsorbed Hg(II) ion concentration (mg/L), while K_L stands for constant (binding affinity).

The isotherm shape can be investigated using the separation factor, or R_L , by using the following equation:

$$R_L = \frac{1}{1 + K_L C_i} \quad (3)$$

K_L (L/mg) is the Langmuir constant, and C_o is the initial Hg(II) ion concentration (mg/L).

The multilayer adsorption mechanism of adsorption on a heterogeneous surface that is, on various active site types with various potentials is explained by the Freundlich isotherm. Equation (4) provides the expression for the Freundlich equation:

$$q_e = (K_F) (C_e^{1/n}) \quad (4)$$

$$\ln q_e = \ln K_F + \frac{1}{n} \ln C_e \quad (5)$$

K_F and n are the Freundlich constants; q_e (mg/g) is the amount adsorbed at equilibrium; C_e (mg/L) is the equilibrium concentration of Hg(II) ions. The Langmuir isotherm's analysis factor data values, $q_{\max}$ (mg/g), K_L , R_L , and R_2 , are determined by graphing a curve C_e/q_e vs C_e for the VG-assisted removal of Hg(II) ions. In respect to the adsorption of Hg(II) ions, a correlation coefficient (R_2) of 0.9998 was found. The measured values (q_e) and the estimated value ($q_{\max}$) coincided, indicating a suitable Langmuir model. The shape of the isotherm is stated to be determined by the value of R_L , which can be either irreversible ($R_L = 0$), favorable ($0 < R_L < 1$), linear ($R_L = 1$), or unfavorable ($R_L > 1$). Defining the adsorption processes is advantageous since the value of R_L for VG is more than 0 and less than 1. Plotting between $\ln q_e$ and $\ln C_e$ was done in accordance with equation 5 to get the Freundlich isotherm factors (n and K_F). Table 2 presents the n value derived from the graph, K_F derived from the plot's intercepts, and the value

of the R^2 correlation coefficient. where the adsorption capacity on the adsorbent's heterogeneous surface is represented by the reciprocal of n . An increasingly heterogeneous adsorbent surface will result from a data value of $1/n$ that is closer to 0. Additionally, the linearity regression coefficients closer to 1 ($R^2 = 0.994$) for the Freundlich model for Hg(II) ion adsorption appear to be in favor. The data value is less than 1.0, indicating that the Freundlich model is not supported by the Freundlich factor n .

Table 2. Results of adsorption isotherm models, Langmuir and Freundlich constants for Hg(II) ions on VG .

Adsorption isotherm	Constant parameters	Values
Langmuir isotherm	$q_{\max}(\text{mg/g})$	87.70
	K_L	0.0229
	R_L	0.3009
	R^2	0.9998
Freundlich isotherm	$K_F (\text{L/mg})^{1/n}$	0.4249
	n	1.1290
	R^2	0.9941

4.7 Study of kinetic models

Studies on kinetics investigate the rate of adsorption processes and elucidate the mechanism. In order to understand the behavior of adsorption, two kinetic models are examined here. Because of this, the adsorption kinetics of Hg(II) ions have been understood by applying pseudo-first-order and pseudo-second-order models to the experimental and theoretical data. For the pseudo-first-order and pseudo-second-order models, the following is Lagergren's equation [20–21]:

$$\log(q_e - q_t) = (\log q_e) - \left(\frac{k_1}{2.303} \right) t \quad (6)$$

$$\frac{t}{q_t} = \left(\frac{1}{k_2 q_e^2} \right) + \left(\frac{1}{q_e} \right) t \quad (7)$$

where q_e is the quantity of Hg(II) ions adsorbed per unit weight of adsorbent at equilibrium at the adsorption constant, k_1 , and q_t (mg/g) is the amount of Hg(II) ions adsorbed at any given

time. For the removal of Hg(II) ions, $\log(q_e - q_t)$ vs time is plotted to get the adsorption constant k_1 . Table 3 presents an overview of the analysis findings for correlation coefficients (R^2) and k_1 . Table 3 demonstrates that the values determined analytically and the experimental values (q_{exp}) coincided. Therefore, correlation coefficient (R^2) values are not toward linearity and the pseudo-first-order model is not appropriate for the adsorption of Hg(II) ions onto VG.

Plotting t/q_t against time allowed for the analysis of the pseudo-second-order model. In the direction of linearity, the slope and intercept of the plot yield the values of q_e (mg/g) and k_2 (g/mg.min). It has been found that the pseudo-second-order model's regression coefficient value is 0.9999. Table 3 indicates a higher degree of agreement between the computed values of q_e and the experimental data. The adsorption curves of Hg(II) ions onto VG are in favor of the pseudo second-order kinetic model, as per the data analysis.

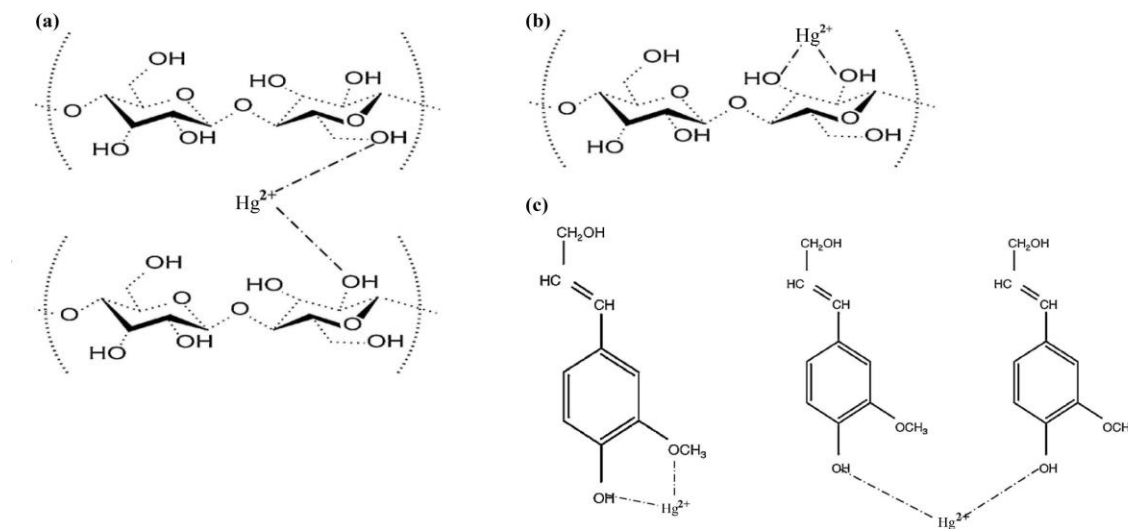
Table 3. Pseudo-first order and pseudo-second-order Kinetic models for the removal of Hg(II) ions on VG.

	Constant parameters	Values	R^2
Kinetics model			
Pseudo-first- order	q_e cal. (mg/g)	152.7	0.5680
	q_e exp. (mg/g)	3.29	
	K_1 (min ⁻¹)	0.0001	
Pseudo-second-order	q_e cal. (mg/g)	153.3	0.9999
	q_e exp. (mg/g)	152.7	
	K^2 (min ⁻¹)	0.8467	

4.1 Hg (II) adsorption mechanism

Adsorbed Hg(II) ions onto VG adsorbent are supported by the FTIR analysis, which shows significant shifts in the spectra peaks, namely O–H around the wavenumber of 3599.01 and 3426.05 cm⁻¹, shifting to 3546.80 and 3432 cm⁻¹, respectively. The peak of NH at 1639.60 cm⁻¹ was slightly moved to 1642 cm⁻¹ due to the presence of mercury ions. Because of the Hg(II) ions, the peaks at 1556 cm⁻¹ and the peak of the –C–N bending vibrations at 1250 cm⁻¹ are moved to 1549.80 cm⁻¹ and 1304 cm⁻¹, respectively. According to Scheme 1, complexation and ion

exchange, both are responsible for the Hg(II) ions suggested adsorption mechanism on the surface of VG adsorbent.



Scheme 1: Proposed binding mechanism in VG (a, b) Cellulose and (c) Lignin with Hg^{2+}

5. Conclusions

It took two hours to determine the maximal adsorption capabilities for Hg^{2+} onto the VG. Since solute adsorption capability is optimum at pH 5, the pH of the solute solution is a crucial parameter. According to an analysis of the isotherm models, the results of the Langmuir isotherm model have a stronger correlation with the R^2 value than the Freundlich isotherm model. The primary functional group on the surface of VG, the carboxylic group, significantly affects the adsorption properties of Hg^{2+} , according to FTIR study as well. The clear variations in peak frequencies were seen in the samples' FTIR spectroscopy data. Due to its status as an agricultural waste, VG is a particularly advantageous material to use for environmental contamination removal. By doing so, the notion of sustainable development will be implemented.

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