

Research Article

Removal of Chromium (VI) and Lead (II) from Aqueous Solution Using Domestic Rice Husk Ash- (RHA-) Based Zeolite Faujasite

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Received 6 June 2022; Accepted 13 July 2022; Published 31 July 2022

Academic Editor: Randa Khalifa

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Rice husk ash (RHA), is a widely available biobased source for high purity silica. In this work, zeolite Faujasite (FAU) is synthesized using extracted silica from RHA (collected from a local region of Bangladesh). The synthesized zeolite FAU was used as an adsorbent for Cr(VI) and Pb (II) batch-wise adsorptive removal from respective aqueous solution. The synthesized zeolite FAU was characterized using X-ray diffraction (XRD), scanning electron microscopy (SEM), nitrogen-sorption, and Fourier transfer infrared (FT-IR) spectroscopy. Metal ion adsorption studies were performed by varying metal concentration (20–100 mg/L for Cr(VI) and 900–133 mg/L for Pb(II)), sorbent dosage (2–10 g/L for chromium and 0.5–1.5 g/L for lead), and contact time (10–120 min for both metal ions). The maximum adsorption capacity of RHA-based zeolite FAU was found to be 3.56 mg/g and 342.16 mg/g for Cr(VI) and Pb(II), respectively. Since the sorption data was found to match with Langmuir isotherm, a monolayer adsorption was occurred. The regeneration of the RHA-based zeolite FAU by NaCl solution showed the potential of repeated as well as continuous operation.

1. Introduction

Water contamination by heavy metals is a major concern for the environment and human health. Heavy metals such as chromium, lead, cadmium, mercury, nickel, copper, and zinc are nonbiodegradable and accumulate in the human body, posing health risks due to their toxicity [1]. Hence, it is necessary to treat metal-contaminated wastewater before its discharge into the environment. Chromium and lead are two of the most hazardous heavy metals for living beings and the environment. Various processes such as mining, leather tanning, electroplating, wood preservatives, paints, and textile dyeing deal with inorganic chemicals and pigments that discharge chromium and lead-based compounds as industrial wastewater [2]. Since Bangladesh is one of the leading textile product producers and has a considerable electroplating demand, contamination of chromium and lead in water is a growing concern here in this country.

From toxicological data, it is obvious that the degree of toxicity depends on the chemical form in which the element is present [3]. In the case of chromium, the trivalent form Cr(III) and the hexavalent form Cr(VI) are more prevalent in the environment and, therefore, are the two main chromium components of concern in respect to water pollution [4]. Cr(III) is supposed to be an essential micronutrient for human, plant, and animal metabolism. On the other hand, Cr(VI) is soluble in a wider pH range and has greater mobility and more toxic than Cr(III). Human can be exposed to excessive amount of Cr(VI) through different media such as food, drinking water, and inhaling air. The respiratory tract is supposed to be the major target organ for Cr(VI). For acute exposure of Cr(VI) coughing, breathing problem and wheezing can occur while for chronic exposure bronchitis, pneumonia, perforations, and ulceration of septum may happen. Chromium(VI) can be heavily lethal if the concentration reaches to 0.1 mg/g of a human weight [5]. Hexavalent

chromium (Cr(VI)) is a probable carcinogen that causes primary liver cancer. Moreover, Cr(VI) belongs to water hazard class 3, which means extremely hazardous. Hence, the maximum permitted concentration in wastewater for hexavalent chromium is $0.05 \text{ mg}\cdot\text{dm}^{-3}$, which is ten times lower than the trivalent form ($5 \text{ mg}\cdot\text{dm}^{-3}$) [6].

In a similar fashion, lead contaminates groundwater through its usage in, for example, batteries, glass ceramics, mining, smelting, and plating industries [7] and in contact with biotic communities causes health problems. When lead builds up in a human body for a longer period (over months or years), lead poisoning can occur. Serious health problems may occur due to even a small amount of lead poisoning. Children are reported to be more vulnerable than adults to lead poisoning because lead poisoning may affect mental as well as physical development. Lead accumulates in muscles, bones, kidneys, and brain tissues and can result in anemia, nervous system disorders, and kidney diseases [8]. Therefore, it is extremely required to effectively remove Cr(VI) and Pb(II) from contaminated effluent of various industries.

An adsorption process is a widely reported technique for the removal of various hazardous pollutants from aqueous solution due to the high efficiency [9] and economic viability [10]. It is also commonly used to remove heavy metals from wastewater compared to other techniques like chemical precipitation [11], ion flotation [12], and biological treatment [13]. Biosorbents [14], synthetic adsorbents [15], and bioresource-derived adsorbents [16] have been reported for the removal of such heavy metals from an aqueous solution. Among them, bioresource-derived sorbents for wastewater treatment provide a greener and sustainable alternative.

Bangladesh is the third-highest rice-producing country per annum in the world. According to the data from the Ministry of Agriculture, Bangladesh, in the fiscal year 2020-21, around 38.7 million metric tons (MMT) of rice were cultivated in Bangladesh [17]. The cultivated rice consists of 20 wt.% rice husk which composition is as follows: cellulose (50%), lignin (25–30%), silica (15–20%), and moisture (10–15%) [18]. Therefore, 1.6 MMT silica was produced in the fiscal year 2020-21 in Bangladesh. Typically, rice husk is used as burning fuel in the rice boiling process, and at the end, rice husk ash (RHA) is left over. Although such a burning process is an uncontrolled one, still around 60% of silica from RHA can be chemically extracted from it [19]. The extracted silica can be used in cement as additives, or as other forms of silica, or can be converted into other materials such as zeolites.

Zeolites are a porous aluminosilicate material. And silicon is an essential structural element for this material. Zeolite Faujasite-type is widely used as ion-exchange material due to its thermochemical stability, being easily tunable and its selective sorption properties, synthesizability using inexpensive elements, and environmental compatibility [20–22]. The inside cages or surface of 7.2 nm sized pore of zeolite FAU acts as sorption active sites for ion species due to the presence of positive aluminum in the framework [22]. Moreover, after completion of adsorption cycle, the zeolite can be regenerated with a simple ion exchange and be used for several sorption cycles resulting in longer service life.

Therefore, in this paper, an eco-friendly and sustainable pathway for removing Cr(VI) and Pb(II) from wastewater using zeolite FAU as adsorbent, which is synthesized from the local bio-sourced of RHA, is presented. The experimental work includes extraction of silica from locally collected waste RHA, its subsequent utilization in zeolite FAU synthesis, and batch-wise adsorptive removal of Cr(VI) and Pb(II) from aqueous solution.

2. Materials and Methods

2.1. Chemicals. Rice Husk Ash was collected from Khan Agro-Rice mill, Muktagachha, Mymensingh, Bangladesh. Other chemicals used were sodium aluminate (43-44% Al_2O_3 , Sigma-Aldrich, Germany), potassium dichromate (99%, Sigma-Aldrich, USA), 1,5-diphenylcarbazide (99%, Sigma-Aldrich, USA), NaOH pellets (97%, Merck, India), H_2SO_4 (98% purity, Merck, India), and HCl (37%, Merck India).

2.2. Silica Extraction. Before silica extraction, RHA was washed with adequate distilled water until discarded water pH was at 7-8 and oven-dried. In a typical silica extraction process, 20 g of washed RHA was mixed with 120 mL 1 N NaOH and left for 5 hours with constant stirring at 75°C . Next, the filtrates (aqueous parts) were collected via filtration. The collected aqueous solution was allowed to cool to room temperature and titrated with adequate 1 N HCl with constant stirring until a gel was formed (typically at $\text{pH} < 8$). The obtained gels were aged for 18 h in room temperature, and by adding deionized water, a slurry was produced. Finally, the formed silica was separated via vacuum filtration and dried in an oven.

2.3. Zeolite FAU Synthesis. The zeolite FAU was synthesized hydrothermally using the seed gel method. First, a seed gel having a molar composition of $10.67 \text{ Na}_2\text{O} : 1 \text{ Al}_2\text{O}_3 : 10 \text{ SiO}_2 : 180 \text{ H}_2\text{O}$ was prepared. Typically, 1.2 g of NaOH pellet was dissolved in 3.43 g of H_2O , and then 6.24 g of sodium metasilicate was added under stirring. The formed seed gel was aged for 18 hr.

For synthesis, a mixer having a molar composition of $4.3 \text{ Na}_2\text{O} : 1 \text{ Al}_2\text{O}_3 : 10 \text{ SiO}_2 : 180 \text{ H}_2\text{O}$ was prepared. First, 4.97 g of NaOH was dissolved in 35.51 g of H_2O under stirring for 15 minutes, and then 3.05 g of NaAlO_2 was added to the mixture and stirred for 30 minutes (mixture 1). In another beaker, an amount of 11.50 g of an extracted silica source was completely dissolved in 35.51 g of water via 15 minutes of stirring, and the resulted solution was then transferred into mixture 1. Finally, previously prepared seed gel was added to the fresh mixture in a mass ratio of 1 (seed gel): 9 (fresh mixture) and stirred for 2 hr. The resulting mixture was put into a polypropylene PP bottle and inserted into a preheated oven at 70°C for 48 hr. After the synthesis, the powder was collected via vacuum filtration and washed with abundant water.

2.4. Cr(VI) and Pb(II) Adsorption Study and Analysis. All batch Cr(VI) and Pb(II) adsorption studies conducted at pH 5 as point of zero charge of synthesized zeolite FAU were

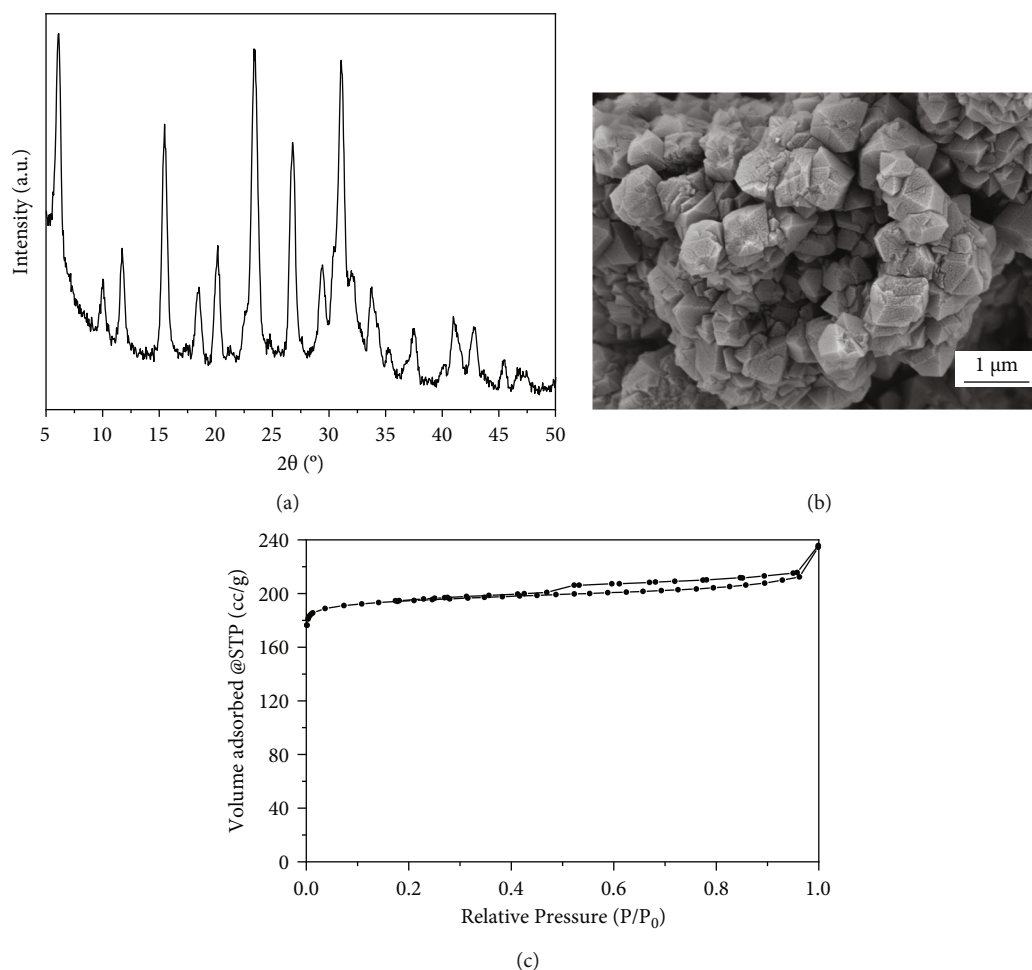


FIGURE 1: Textural properties of RHA silica-based zeolite FAU: (a) XRD pattern, (b) SEM image, and (c) N_2 -sorption.

found to be pH 7.3 (Figure S1). The starting solutions were prepared by dissolving required potassium di-chromate and lead nitrate in deionized water, respectively. During the adsorption, the solution and adsorbent were put into a 200 rpm shaker for the required time. The Cr(VI) content in the solution was determined via the Di-phenyl carbazide (DPC) method using UV-vis 1900i (Shimadzu) at a detection range of 300-700 nm (Figure S2). Pb(II) content was determined using the AA-7000 (Shimadzu) AAS spectrometer.

2.5. Regeneration of Adsorbent. Regeneration tests of the adsorbent for Cr(VI) and Pb(II) removal was carried out by using sodium chloride (NaCl) solution. After the adsorption test, the adsorbent was collected and washed with NaCl solution to desorb or elute the adsorbed metal ions. Thus, the active sites of the adsorbent became unoccupied and the adsorbent became ready for adsorption again. Then, the adsorption experiment was done in the same fashion as before.

3. Characterization

XRD analysis was carried out using an Empyrean diffractometer (Malvern Panalytical) in the 2θ range of 5-90°.

SEM analysis was done using GeminiSEM 500 (Zeiss); N_2 -Sorption was carried out at -196°C in a Quadrasorb™ SI gas sorption analyzed after the sample was pretreated at 250°C in a vacuum for 12 h. FT-IR analysis was carried out in the Nicolet iS20 FTIR spectrometer.

4. Results and Discussion

4.1. RHA-Based Zeolite FAU Textural Properties. XRD pattern (Figure 1(a)) of synthesized powders shows prominent peaks at 2θ positions of 6.1°, 10.2°, 11.7°, 15.6°, 18.4°, 20.2°, 23.6°, 26.8°, 31.2°, and 33.9°, which match with zeolite Faujasite crystallography patterns. Scanning electron microscopy (SEM) images provide an insight into the morphology of the studied material. Figure 1(b) represents the SEM image of RHA-based zeolite FAU. It shows that the surface of the zeolite consists of an inhomogeneous crystalline structure (octahedral shape) with a particle size of around 1 μm . N_2 -sorption analysis shows (Figure 1(c)) IUPAC type I isotherms, which indicates microporous material. The synthesized zeolite FAU has a specific surface area of 661.2 m^2/g .

4.2. Cr(VI) and Pb(II) Adsorption on RHA-Based Zeolite FAU. To know the insight into the metal ion sorption

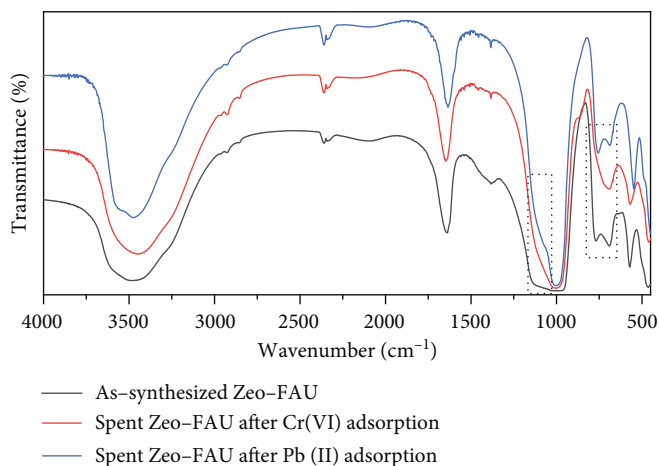


FIGURE 2: FT-IR spectra of synthesized RHA-based zeolite FAU before and after adsorption of Pb(II) and Cr(VI) ions.

mechanism by RHA-based zeolite FAU, FT-IR is a useful tool for material characterization. Figure 2 shows the FT-IR spectra of synthesized RHA-based zeolite FAU before and after adsorption of Pb(II) and Cr(VI) ions over a range of 400–4000 cm^{-1} . In the case of RHA-based zeolite FAU before adsorption of metal ions, strong broadband at 3500 cm^{-1} was observed. This band indicated O-H stretching as well as Si-OH and Si-OH-Al. At 1640 cm^{-1} , a strong band was spotted which was supposed to be assigned for O-H bending vibration of adsorbed metal ion onto the adsorbent. A wide and broader band in the range 950–1150 cm^{-1} was associated with Si-O-Al asymmetric stretching vibration. A double band around 685–770 cm^{-1} was attributed to the symmetric stretching vibration of Si-O-Si or Si-O-Al. A sharp band at 570 cm^{-1} was due to external linkage vibration, which is connected to sodalite cages (Si-O-Na). The comparison of spectra showed that characteristic peaks of RHA-based zeolite FAU have been slightly shifted toward high or low wavenumber after adsorption of metal ions. This might happen due to the introduction of cations into the framework of the adsorbent because the interaction between the adsorbent and the metal ion could change the IR spectra [23]. As in the case of Pb(II) adsorption, a narrow peak at 983 cm^{-1} was observed. This peak originated from Pb-O vibrations [7]. On the other hand, a double band disappeared and a single band at 686 cm^{-1} was observed. This might happen due to the adsorption of Cr(VI) by the zeolite.

4.3. Adsorption Parameters Study

4.3.1. Effect of Contact Time. Determining the optimum agitation time for obtaining better sorption performance is one of the key issues in the adsorption process. In this study, the percentage removal of chromium as well as lead using RHA-based zeolite FAU was investigated with varying agitation time (such as 10, 20, 40, 60, 90, and 120 min). Agitation speed and temperature were kept constant for both experiments. As depicted in Figure 3, the percent removal increases with increasing contact time. At the initial stage of the tests, the adsorption process was quite fast and maxi-

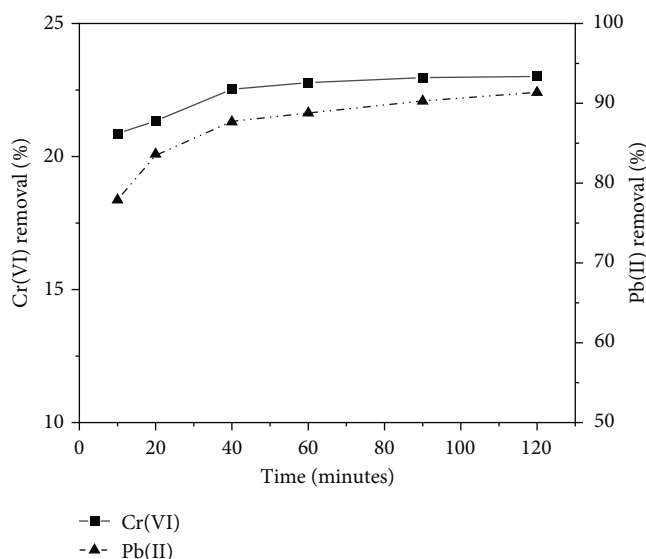


FIGURE 3: Removal of chromium and lead as a function of time by using RHA-based zeolite FAU.

imum removal of metal ions occurred in 2 h. The figure shows that 88% lead removal occurred within 40 minutes, whereafter a very little lead removal (from 88% to 91%) was observed. However, equilibrium was achieved in 2 h time. A similar trend of adsorption was also observed for the adsorptive removal of chromium from an aqueous solution (Figure 3). Therefore, 2 h was selected as the optimum time for the adsorption process for the removal of both metal ions.

At the beginning of the adsorption process, there were a large number of active sites available to take part in the adsorption process. That is the reason for the faster adsorption of the metal ion by the adsorbent. As the process goes on, the active binding sites become gradually occupied by the metal ion and the removal process slows down. Hence, fraction lessening of adsorbent surface and competition among metal ions are supposed to be responsible for decreasing metal ion removal. Similar findings were reported

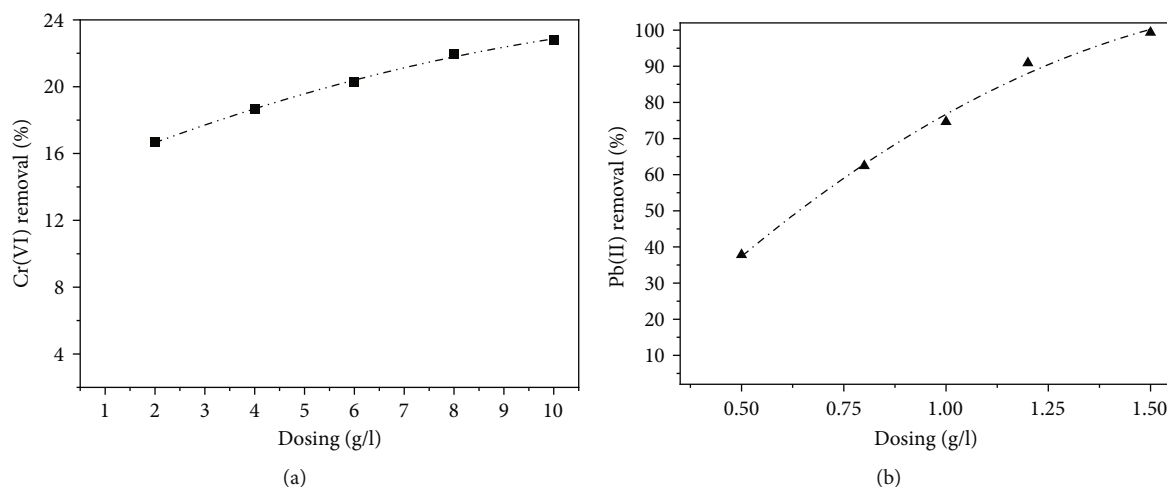


FIGURE 4: Effect of adsorbent dosing on the removal of (a) Cr(VI) and (b) Pb(II) from aqueous solution by using RHA-based zeolite FAU.

by Khalil et al. and Badessa et al. [24, 25] who removed Cr(VI) from aqueous solution by using different biobased sorbents.

4.3.2. Effect of Adsorbent Dosing on Cr(VI) and Pb(II) Removal. The adsorbent dose is one of the key parameters that should be known to select optimal conditions for the adsorption process. The variations of adsorbent dose were tested in this study by keeping adsorbate concentration, pH, agitation speed, and operating temperature constant. Figures 4(a) and 4(b) show the effect of the adsorbent dose for removal of Cr(VI) and Pb(II), respectively, by using the same adsorbent, namely, rice husk ash-based zeolite FAU. The percent of metal removal through adsorption was found to improve with rising adsorbent doses. Figures 4(a) and 4(b) show that chromium removal by RHA-based zeolite FAU rises from 17% to 23% for a rise in dosing from 2 g/L to 10 g/L. In the case of lead, the percent removal increases from 38% to almost 100% for a rise in adsorbent dose from 0.5 g/L to 1.5 g/L.

Because of the increase in adsorbent dose, the surface area of the adsorbent enhances resulting in increased binding sites for more metal ions to be adsorbed [26]. After a certain dose, the adsorption process reaches equilibrium whereafter the percent removal of metal ions is no more affected by the increase in dose. A similar pattern of dosing influence on the removal of lead from aqueous solution by using combination of oil shale ash and coal fly ash-derived zeolite FAU (X-type) was reported in the literature [27].

4.3.3. Effect of Metal Ion Concentration. The initial concentration of adsorbate is considered a crucial parameter because it provides the necessary driving force for the transport of adsorbate from the bulk solution to the surface of the adsorbent [24]. The adsorption capacity of RHA-zeolite was studied separately for the adsorption of chromium as well as for lead by varying initial adsorbate concentration and by keeping all other parameters (e.g. pH, agitation time, agitation speed, adsorbent dosage, and temperature) constant. Initial metal ion concentration for chromium was spanning

from 20 to 100 mg/L while that for the lead was 900 to 1300 mg/L. For both metals, it was observed that when the metal ion concentration is low, the adsorption capacity of the adsorbent was at its minimum. But with the increase in metal ion level, the adsorptive process between the sorption sites and metal ions increased, which enhanced the metal ion uptake until an adsorption equilibrium was attained. The effect of metal ion concentration on adsorbent capacity has been shown in Figure 5 from where it is obvious that with the increase in ion concentrations (for both lead and chromium), the adsorption capacity increases. This phenomenon can be explained by the fact that all available active sites of the RHA-based zeolite FAU would be completely exposed to get filled by the metal ions of the solution [28]. With a further increase in Pb(II) concentration, the adsorption capacity does not significantly increase and eventually forms a plateau from where the maximum adsorption capacity can be determined. From the experimental data of this study, the maximum sorption capacity was determined to be 3.56 mg/g for Cr(VI) and 342.5 mg/g for Pb(II). The extent of adsorption capacity is quite comparable to other similar adsorbents. A comparison has been tabulated below in Table 1.

4.4. Adsorption Isotherm Modeling. When a solid-liquid adsorption system is concerned, it is required to know the isotherm model. The reason is that the adsorption isotherm model necessarily describes the adsorption behavior. The adsorption isotherm specifies the distribution of adsorbate molecules between the solid phase and liquid phase when the system is in equilibrium. Since it is significant to understand the adsorption behavior, the distribution of metal ions between the phases has been determined by two well-known adsorption isotherm models, namely, (i) Langmuir isotherm and (ii) Freundlich isotherm. The modeling has been accomplished by fitting the experimental data into the specific isotherm equations.

4.4.1. Langmuir Isotherm. The assumption of the Langmuir isotherm model includes a feature, which expresses that

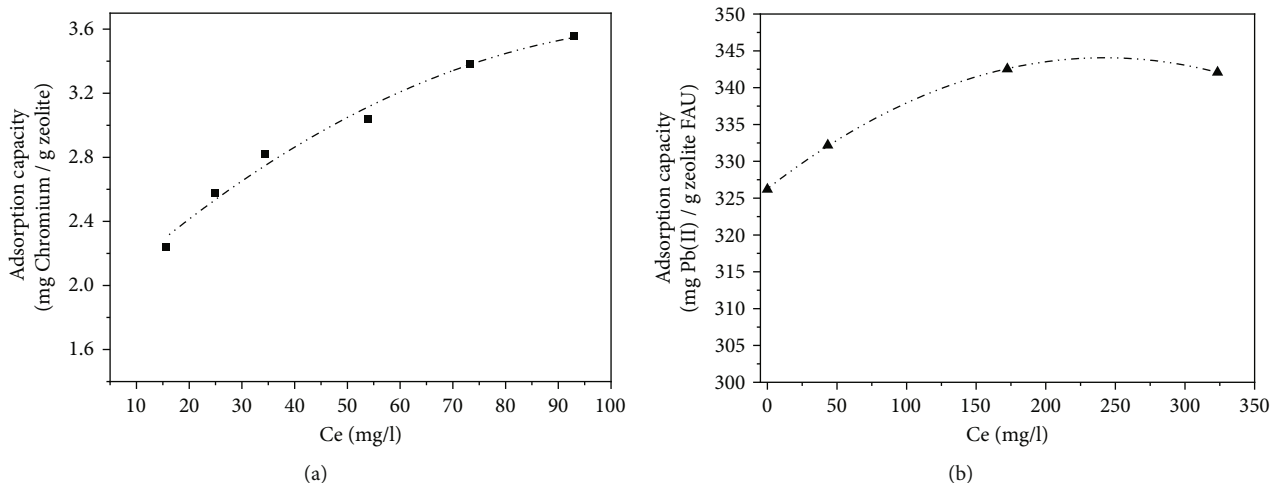


FIGURE 5: Effect of metal ion concentration on the removal of (a) Cr(VI) and (b) Pb(II) from aqueous solution by using RHA-based zeolite FAU.

TABLE 1: Comparison of notable aluminium-rich zeolites for Pb(II) and Cr(VI) removal from aqueous solution.

Adsorbent	Silica source	Adsorbate	Capacity (mg/g)	Reference
Zeolite A	Red mud	Pb (II)	100	29
Zeolite X	Mixed of oil shale ash and coal fly ash	Pb (II) in mixed metal ion solution	112.5	27
Zeolite X	Coal fly ash	Pb (II)	314	30
Zeolite 13 X	Commercial	Cr (VI)	3.92	28
Natural zeolite	Not required	Cr (VI)	3.2	31
Zeolite NaP1	Coal fly ash	Cr(VI)	17.92	32
Zeolite FAU	Rice husk ash	Pb(II)	342.5	This study
Zeolite FAU	Rice husk ash	Cr(VI)	3.56	This study

monolayer adsorption (of the adsorbate molecule) occurs on the homogeneous surface of the adsorbent [33]. Langmuir isotherm can be expressed as the following equation (Equation (1)). This equation can be simplified by mathematical operation and can be rewritten as Equation (2).

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e}, \quad (1)$$

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

where q_e stands for metal ion adsorbed per gram of adsorbent (mg/g), q_m is the maximum adsorption capacity with the same unit of q_e , C_e designates equilibrium concentration of metal ion (mg/L), and K_L indicates Langmuir constant (L/mg).

From the simplified Langmuir equation, a plot of C_e/q_e vs. C_e has been depicted in Figure 6. Considering the value of the correlation coefficient (R^2), the experimental data was found to fit well with the linear Langmuir isotherm equation. The maximum adsorption capacity of RHA-based zeolite FAU and the value of equilibrium constant was calculated from the slope and the intercept of the straight line, respectively. The calculated sorption capacities

were 3.95 mg-Cr(VI)/g and 342.46 mg Pb(II)/g, which were consistent with the experimental data (Tables 1 and 2). Therefore, it is perceived that a monolayer adsorption of metal ions has occurred. A dimensionless parameter, also called equilibrium separation factor (R_L), is considered one of the essential characteristics of the Langmuir isotherm. The equilibrium separation factor can be defined as follows:

$$R_L = \frac{1}{1 + K_L C_0}, \quad (3)$$

where K_L (L/mg) and C_0 (mg/L) are designated as Langmuir constant and initial adsorbate concentration, respectively. The values of R_L were calculated (for both chromium and lead ions) and found to remain between 0 and 1. This means that the adsorption was favorable under the studied conditions.

4.4.2. Freundlich Isotherm. Freundlich isotherm is an empirical mathematical model which represents the amount adsorbed per unit amount of adsorbent. The mathematical model of the Freundlich isotherm is based on the assumptions that both anions and cations can be adsorbed on the same surface simultaneously. It articulates multilayer

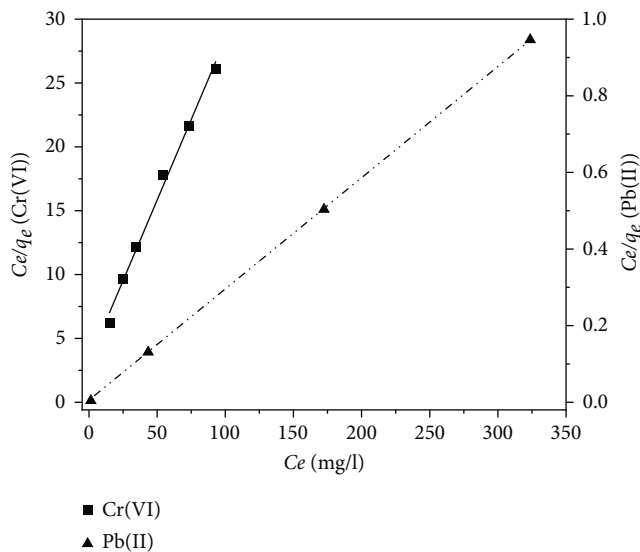


FIGURE 6: Langmuir isotherm plot for the removal of Cr(VI) and Pb(II) by RHA-based zeolite FAU.

TABLE 2: Parametric values of Langmuir and Freundlich isotherm data.

Isotherm model	Parameters	Values	
		Cr(VI)	Pb(II)
Langmuir isotherm	K_L (L/mg)	0.0784	1.7391
	q_m (mg/g)	3.95	342.46
	R^2	0.991	0.999
Freundlich isotherm	K_f (mg/g)	1.3269	324.31
	$1/n$	0.2146	0.0092
	R^2	0.971	0.896

adsorption onto a heterogeneous surface. The Freundlich model can be defined as follows:

$$q_e = K_f C_e^{1/n}, \tag{4}$$

where q_e (mg/g) is the amount of metal ion adsorbed at equilibrium, C_e (mg/L) is the equilibrium concentration of a metal ion in the solution, and K_f and $1/n$ are the Freundlich empirical parameters, which are related to adsorption capacity and adsorption intensity, respectively. The adsorption intensity may vary with the heterogeneity of the adsorbent surface. To determine the essential Freundlich parameters this equation can be made linear. The linearized form of this equation can be represented as follows:

$$\ln q_e = \ln K_f + \frac{1}{n} \ln C_e. \tag{5}$$

The value of K_f and $1/n$ can be evaluated from the intercept and slope of the linear plot of $\ln q_e$ vs. $\ln C_e$. If $0.1 < 1/n < 1$ and if K_f is larger, the adsorption system

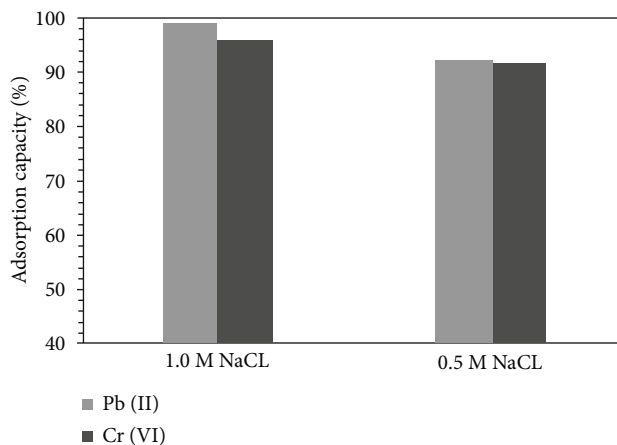


FIGURE 7: Regeneration of adsorbent with different concentration sodium chloride solution.

is thought to be favorable. The parameters obtained from Freundlich isotherm model have been presented in Table 2. It is seen from Table 2 that the value of K_f is small for the removal of Cr(VI) and the value of $1/n$ is smaller than 0.1 for Pb(II) adsorption. Therefore, neither of the metal ion adsorption follows the Freundlich isotherm.

4.5. Regeneration and Economical Analysis. Although adsorption is widely used for remediation of water contamination, often, it cannot be used for commercial utilization because of its disposal problem as well as its incapability to reuse [34]. Therefore, the efficiency of regeneration of adsorbents is something that should be focused on. Moreover, regeneration is supposed to be an important feature because it controls the economy of the water treatment process to a huge extent. For the sake of material cycling, it is needed to remove the adsorbed metal ion from the metal ion-loaded adsorbent. Through the desorption process, the metal ion can be removed from the active binding sites of the adsorbent surface and thus the adsorbent becomes ready for further adsorption. This process (desorption) can also be termed as elution, where an appropriate chemical reagent (called eluent) can be used to desorb the metal ions that are already loaded on the adsorbent surface. For effective regeneration of adsorbent as well as heavy metal recovery, various acids, alkalis, complexing agents, salts, etc. are used [34, 35]. The choice of eluent largely depends on binding chemistry between adsorbent and metal ion, endurance, and tolerance of adsorbents towards the reagents, and so forth.

In this study, regeneration experiments were conducted by using one of the most abundant minerals on the earth, NaCl, rather than using acids and alkalis. Sodium chloride was chosen for this purpose to avoid acid-base consumption. Salt (NaCl) concentrations of 1.0 M and 0.5 M were used for regenerating the adsorbent. The results are presented in Figure 7, which shows that 99% and 92% regeneration of the RHA-based zeolite FAU adsorbent (in case of Pb(II) removal) was possible by using NaCl of 1.0M and 0.5M, respectively. Similarly, 95% and 91% regeneration of the RHA-based zeolite FAU (for Cr(VI) removal) was found to

occur by using sodium chloride concentrations of 1.0 M and 0.5 M, respectively. The result indicated that regeneration of rice husk ash-based zeolite FAU adsorbent is very much possible by using an inorganic chloride salt, namely, sodium chloride.

The economical perspective of any process is a key factor [36] to consider. The commercial zeolite FAU is relatively cheap materials costing around \$ 0.3-0.4 per kilogram. As one of the building elements (silica) of the synthesized zeolite FAU in this study was extracted from the relatively abundant and cheap materials (rice husk), the final cost will be lower than those of commercial ones. The high sorption capacity of regenerated adsorbent (99% and 92% for Pb(II) and Cr(VI), respectively) makes the material economically more attractive.

5. Conclusion

Zeolite Faujasite was successfully prepared from an agroindustrial waste precursor, namely, rice husk ash. It was applied for the removal of pollutants (lead and nickel ions) from contaminated water. Batch-wise experiments were conducted to establish the most favorable conditions for Pb(II) and Cr(VI) removal. SEM image exhibited the morphology of the adsorbent where a crystalline structured octahedral shape was observed. From BET surface analysis, the surface area of the adsorbent was found to be 661.2 m²/g. Characterization of the adsorbent displayed that there was change in FTIR spectra before and after adsorption of metal ions, which was the proof of metal ion adsorption by rice husk ash-based zeolite-FAU as sorbent. The percentage removal of Pb(II) as well as Cr(VI) ion increased with the increase in adsorbent dose and contact time. During an adsorption process, equilibrium was achieved in 120 min. The adsorption process of Cr(VI) and Pb(II) followed the Langmuir isotherm model. Maximum sorption capacity of the adsorbent was 3.56 mg-Cr(VI)/g and 342.5 mg-Pb(II)/g. Regeneration, using different concentration of NaCl solution, indicated a potency to reuse the RHA-based zeolite-FAU. This process could be well-integrated into a waste treatment plant where the water is contaminated with heavy metals like Pb(II) and Cr(VI).

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declared there are no conflicts of interest with respect to the research, publication of this article, and authorship.

Acknowledgments

The authors are grateful to Jashore University of Science and Technology for granting the research fund.

Supplementary Materials

Supplementary 1. Figure S1 exhibits the point of zero charge (pH_{pzc}) of the surface of the adsorbent (RHA-based zeolite FAU). The pH_{pzc} is the pH at which the net surface charge of the adsorbent is zero. The extent of pH_{pzc} was found to be 7.3 in this study. The concept of pH_{pzc} has drawn much attention because the adsorption of many substances is extremely dependent on the solution pH.

Supplementary 2. Figure S2 demonstrates a calibration curve for Cr(VI) detection via UV-vis DPC method. A detailed detection process was provided in the supplementary data. The sample was analyzed at 300-800 nm range UV-vis spectroscopy (a peak at 553 nm).

References

- [1] R. Vinoth Kumar and G. Pugazhenth, "Removal of chromium from synthetic wastewater using MFI zeolite membrane supported on inexpensive tubular ceramic substrate," *Journal of Water Reuse and Desalination*, vol. 7, no. 3, pp. 365-377, 2017.
- [2] T. Ai, X. Jiang, and Q. Liu, "Chromium removal from industrial wastewater using phyllostachys pubescens biomass loaded Cu-S nanospheres," *Open Chemistry*, vol. 16, no. 1, pp. 842-852, 2018.
- [3] L. Rafati, A. H. Mahvi, A. R. Asgari, and S. S. Hosseini, "Removal of chromium (VI) from aqueous solutions using lewatif fo36 nano ion exchange resin," *International journal of Environmental Science and Technology*, vol. 7, no. 1, pp. 147-156, 2010.
- [4] R. Leyva-Ramos, A. Jacobo-Azuara, P. E. Diaz-Flores, R. M. Guerrero-Coronado, J. Mendoza-Barron, and M. S. Berber-Mendoza, "Adsorption of chromium(VI) from an aqueous solution on a surfactant-modified zeolite," *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, vol. 330, no. 1, pp. 35-41, 2008.
- [5] B. Kakavandi, R. R. Kalantary, M. Farzadkia et al., "Enhanced chromium (VI) removal using activated carbon modified by zero valent iron and silver bimetallic nanoparticles," *Journal Of Environmental Health Science and Engineering*, vol. 12, pp. 1-10, 2014.
- [6] M. Pang, N. Kano, and H. Imaizumi, "Adsorption of chromium (VI) from aqueous solution using zeolite/chitosan hybrid composite," *J. Chem. Chem. Eng.*, vol. 9, no. 7, pp. 433-441, 2015.
- [7] N. Bu, X. Liu, S. Song et al., "Synthesis of NaY zeolite from coal gangue and its characterization for lead removal from aqueous solution," *Advanced Powder Technology*, vol. 31, no. 7, pp. 2699-2710, 2020.
- [8] S. A. Kim, S. Kamala-Kannan, K. J. Lee et al., "Removal of Pb(II) from aqueous solution by a zeolite-nanoscale zero-valent iron composite," *Chemical Engineering Journal*, vol. 217, pp. 54-60, 2013.
- [9] S. Soni, P. K. Bajpai, J. Mittal, and C. Arora, "Utilisation of cobalt doped iron based MOF for enhanced removal and recovery of methylene blue dye from waste water," *Journal of Molecular Liquids*, vol. 314, p. 113642, 2020.
- [10] A. Patel, S. Soni, J. Mittal, A. Mittal, and C. Arora, "Sequestration of crystal violet from aqueous solution using ash of black turmeric rhizome," *Desalination and Water Treatment*, vol. 220, pp. 342-352, 2021.

- [11] A. Pohl, "Removal of heavy metal ions from water and wastewaters by sulfur-containing precipitation agents," *Water, Air, and Soil Pollution*, vol. 231, no. 10, 2020.
- [12] M. Taseidifar, F. Makavipour, R. M. Pashley, and A. F. M. M. Rahman, "Removal of heavy metal ions from water using ion flotation," *Environmental Technology and Innovation*, vol. 8, pp. 182–190, 2017.
- [13] A. M. Hamdan, H. Abd-El-Mageed, and N. Ghanem, "Biological treatment of hazardous heavy metals by *Streptomyces rochei* ANH for sustainable water management in agriculture," *Scientific Reports*, vol. 11, no. 1, pp. 1–12, 2021.
- [14] B. R. Chanda, A. H. Mithun, M. A. Hasan, and B. K. Biswas, "Nickel removal from aqueous solution using chemically treated mahogany sawdust as biosorbent," *Journal of Chemistry*, vol. 2021, pp. 1–10, 2021.
- [15] X. Zhang, J. Gao, S. Zhao et al., "Hexavalent chromium removal from aqueous solution by adsorption on modified zeolites coated with Mg-layered double hydroxides," *Environmental Science and Pollution Research*, vol. 26, no. 32, pp. 32928–32941, 2019.
- [16] I. V. Joseph, L. Tosheva, and A. M. Doyle, "Simultaneous removal of Cd(II), Co(II), Cu(II), Pb(II), and Zn(II) ions from aqueous solutions via adsorption on FAU-type zeolites prepared from coal fly ash," *Journal of Environmental Chemical Engineering*, vol. 8, no. 4, p. 103895, 2020.
- [17] U. S. Agriculture, "Department of Bangladesh: Grain and Feed Annual," <https://www.fas.usda.gov/data/bangladesh-grain-and-feed-annual-6>.
- [18] B. Singh, "Rice husk ash," in *Waste and Supplementary Cementitious Materials in Concrete*, Woodhead Publishing, 2018.
- [19] K. Kordatos, S. Gavela, A. Ntziouni, K. N. Pistiolas, A. Kyritsi, and V. Kasselouri-Rigopoulou, "Synthesis of highly siliceous ZSM-5 zeolite using silica from rice husk ash," *Microporous and Mesoporous Materials*, vol. 115, no. 1-2, pp. 189–196, 2008.
- [20] C. S. Cundy and P. Cox, "The hydrothermal synthesis of zeolites: Precursors, intermediates and reaction mechanism," *Microporous Mesoporous Mater*, vol. 82, pp. 1–78, 2005.
- [21] R. Chanda, L. Wang, and W. Schwieger, "Heat and mass transfer properties of zeolite coatings: comparison of reactive- and spray-coated systems," *Chemie Ingenieur Technik*, vol. 90, no. 5, pp. 708–712, 2018.
- [22] L. F. De Magalhães, G. R. Da Silva, and A. E. C. Peres, "Zeolite application in wastewater treatment," *Adsorption Science and Technology*, vol. 2022, pp. 1–26, 2022.
- [23] M. Król, W. Mozgawa, W. Jastrzębski, and K. Barczyk, "Application of IR spectra in the studies of zeolites from D4R and D6R structural groups," *Microporous and Mesoporous Materials*, vol. 156, pp. 181–188, 2012.
- [24] U. Khalil, M. B. Shakoob, S. Ali, and M. Rizwan, "Tea waste as a potential biowaste for removal of hexavalent chromium from wastewater: equilibrium and kinetic studies," *Arabian Journal of Geosciences*, vol. 11, no. 19, p. 573, 2018.
- [25] T. S. Badessa, E. Wakuma, and A. M. Yimer, "Bio-sorption for effective removal of chromium(VI) from wastewater using *Moringa stenopetala* seed powder (MSSP) and banana peel powder (BPP)," *BMC Chemistry*, vol. 14, no. 1, p. 71, 2020.
- [26] D. Song, K. Pan, A. Tariq et al., "Adsorptive removal of toxic chromium from waste-water using wheat straw and *Eupatorium adenophorum*," *PLoS One*, vol. 11, no. 12, article e0167037, 2016.
- [27] S. Bai, M. Chu, L. Zhou, Z. Chang, C. Zhang, and B. Liu, "Removal of heavy metals from aqueous solutions by X-type zeolite prepared from combination of oil shale ash and coal fly ash," *Energy Sources, Part A: Recovery, Utilization, and Environmental Effects*, pp. 1–11, 2019.
- [28] Y. Jin, Y. Wu, J. Cao, and Y. Wu, "Adsorption behavior of Cr(VI), Ni(II), and Co(II) onto zeolite 13x," *Desalination and Water Treatment*, vol. 54, no. 2, pp. 511–524, 2015.
- [29] W. M. Xie, F. P. Zhou, X. L. Bi et al., "Accelerated crystallization of magnetic 4A-zeolite synthesized from red mud for application in removal of mixed heavy metal ions," *Journal of Hazardous Materials*, vol. 358, pp. 441–449, 2018.
- [30] R. Panek, M. Medykowska, M. Wiśniewska, K. Szewczuk-Karpisz, K. Jędruchiewicz, and M. Franus, "Simultaneous removal of Pb²⁺ and Zn²⁺ heavy metals using fly ash Na-X zeolite and its carbon Na-X(C) composite," *Materials (Basel)*, vol. 14, no. 11, p. 2832, 2021.
- [31] M. Akbari Binabaj, S. M. Nowee, and N. Ramezani, "Comparative study on adsorption of chromium(VI) from industrial wastewater onto nature-derived adsorbents (brown coal and zeolite)," *International journal of Environmental Science and Technology*, vol. 15, no. 7, pp. 1509–1520, 2018.
- [32] Y. Zhang, L. Zhou, L. Chen et al., "Synthesis of zeolite Na-P1 from coal fly ash produced by gasification and its application as adsorbent for removal of Cr(VI) from water," *Frontiers of Chemical Science and Engineering*, vol. 15, no. 3, pp. 518–527, 2021.
- [33] I. Langmuir, "The constitution and fundamental properties of solids and liquids. Part I. Solids," *Journal of the American chemical society*, vol. 38, no. 11, pp. 2221–2295, 1916.
- [34] S. Lata, P. K. Singh, and S. R. Samadder, "Regeneration of adsorbents and recovery of heavy metals: a review," *International journal of Environmental Science and Technology*, vol. 12, no. 4, pp. 1461–1478, 2015.
- [35] B. K. Biswas, J. I. Inoue, K. Inoue et al., "Adsorptive removal of As(V) and As(III) from water by a Zr(IV)-loaded orange waste gel," *Journal of Hazardous Materials*, vol. 154, no. 1-3, pp. 1066–1074, 2008.
- [36] S. Soni, P. K. Bajpai, D. Bharti, J. Mittal, and C. Arora, "Removal of crystal violet from aqueous solution using iron based metal organic framework," *Desalination and Water Treatment*, vol. 205, pp. 386–399, 2020.