The effective Ni(II) removal of red mud modified chitosan from aqueous solution

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Abstract

To remove Ni(II) ions from an aqueous solution, researchers used red mud modified by chitosan (RM/CS) material as a new adsorbent. According to the findings, the surface area of red mud is nearly doubled after being treated with chitosan, from 68.6 m²/g to 105.7 m²/g. The effects of pH solution, contact time, and material dosage on the Ni(II) uptake were examined. In comparison with the pseudo-first-order and pseudo-second-order models, the intra-diffusion model was the most suitable kinetic model for the Ni(II) removal. Besides, the three-parameter Sips model was used to predict the Ni(II) adsorption of RM/CS from aqueous solution. Furthermore, the Langmuir maximum Ni(II) uptake capacity of this material was 31.66 mg/g at 323K, which was higher than red mud and several other natural materials. Notably, thermodynamic investigations demonstrated that Ni(II) adsorption on RM/CS is both exothermic and physisic.

1. Introduction

Nowadays, the development of various industries has produced a tremendous amount of waste containing hazardous and carcinogenic heavy metal ions, including Ni(II) ions. When Ni(II) concentrations in aqueous solution surpass 0.07 mg/L, according to WHO, these ions become hazardous (Çelebi et al. 2020). Scientists have been working to discover effective and reasonably affordable ways to remove heavy metal ions from aqueous solutions due to the rising water pollution problem. Precipitation, reverse osmosis, extraction, and adsorption are some of the approaches that have been used (Bartczak et al. 2018). Because of the reusability of adsorbents via the desorption process, adsorption is one of the most effective ways for removing metal ions from aqueous solution among the approaches as mentioned above (Bartczak et al. 2018).

In recent years, numerous studies have employed wastes from agricultural and industrial operations as adsorbents to limit environmental pollution concerns while also bringing economic advantages to society. Some adsorbents can be used in this process, such as crab shells (Jeon 2011, Pradhan et al. 2005), chitosan (Vakili et al. 2019, Wan Ngah et al. 2011), lignin (Supanchaiyamat et al. 2019), and fly ash (Alinnor 2007). One of the solid wastes in this category of materials is red mud, which is produced during the Bayer process of producing alumina from bauxite ore. Because sodium hydroxide solution is used to decompose the bauxite ore in the alumina manufacturing process⁹, the red mud has a pH range of 10 to 13. The composition of red mud mainly contains Fe₂O₃, Al₂O₃, Na₂O, SiO₂, Na₂O, CaO, TiO₂, and several rare elements (Khairul et al. 2019). These chemical compositions and alkalinity are the primary sources of environmental damage. From 1 to 1.5 tons of red mud are discharged into the environment to create 1 ton of aluminum oxide (Khairul et al. 2019). Annually, the amount of red mud discharged is estimated at nearly 150 million tons (Khairul et al. 2019), resulting in the proper disposal of red mud as a significant challenge for the alumina industries. To deal with red mud thoroughly, red mud has been utilized as an adsorbent to remove pollutants such as heavy metal ions (Apak et al. 1998, Nadaroglu et al. 2010, Soner Altundoğan et al. 2000), dyes (Namasivayam & Arasi 1997, Namasivayam et al. 2001), and various inorganic anions (Çengeloğlu et al. 2002, Pradhan et al. 1998). However, their adsorption

capacity is not high because the characteristics of red mud are significantly depend on their origin or where they are found (Khairul et al. 2019). Various methods have been conducted to modify red mud, such as acid treatment (Sahu et al. 2013, Smičiklas et al. 2014), heat treatment (de Jesus et al. 2015, Yang et al. 2020), and modification by synthetic polymers (Ni et al. 2015). Nonetheless, there have not been a number of studies on altering red mud materials with natural polymers that are both ecologically benign and low-cost.

For the reasons stated above, RM/CS, a novel material, was successfully synthesized via the modification of red mud by chitosan. This material was used to remove Ni(II) metal ions in an aqueous solution. The physicochemical properties of materials were analyzed using modern analytical techniques. The factors affecting the Ni(II) removal of this material were also examined, consisting of pH solution, contact time, and adsorbent dosage. In addition, the adsorption mechanism for Ni(II) ions was also determined based on kinetic, isotherm, and thermodynamic studies.

2. Experimental

2.1. Materials and chemicals

Red mud (RM) used in these experiments is taken from the waste of the Saigon underground water plant, Ho Chi Minh City, Vietnam. Chitosan (flake form, yellow color) was synthesized by a research team from the Institute of Nuclear Research, Da Lat, Vietnam, with the following parameters: moisture content of 12.5%, acetylation degree of 90%, and molecular weight 91.7 kDa. Other chemicals used in these experiments were purchased from Merck, including nickel standard solution 1000 mg/L, acid nitric HNO₃ 63%, sodium hydroxide NaOH 98% and DI water.

2.2. Preparation of RM/CS

Red mud/chitosan material (RM/CS) was prepared according to the procedure shown in Scheme 1. Herein, 10.0 g of red mud was placed to 100 mL of chitosan solution containing 2.0 g of chitosan dissolved in 2% acetic acid solution. Next, the obtained mixture was stirred at 1000 rpm for 24 h to complete the modification. Thereafter, the resulting RM/CS mixture was centrifuged at 5000 rpm, washed with DI water several times before drying at 100 °C for 24 hours.

2.3. Instruments

The surface areas of RM and RM/CS materials were measured by the BET surface area analyzers (Quantachrome NOVA 1000e, USA), whereas the morphologies of these materials were observed using a scanning electron microscope (SEM) (S-4800, Hitachi, Japan).

2.4. Batch adsorption investigation

The standard nickel solution (1000 mg/L) was used to make Ni(II) solutions with defined concentrations. The pH values of this solution were adjusted using 1 M HNO₃ and 1 M NaOH. Next, 0.1 g of RM/CS adsorbent was added into a series of beakers containing 50 mL of the prepared Ni(II) solution. Then, the

adsorption process was performed at constant speed on a 10-point magnetic stirrer (RSM-03-10K, Phoenix Instrument, Germany). After stirring, the solution was filtered and diluted with the appropriate dilution factor. The concentrations of Ni(II) before and after adsorption was analyzed by the AAS method (AAS-6300, Shimadzu, Japan). The adsorption capacity (Q_e) and elimination efficiency (%H) of Ni(II) ions were determined according to the following formulas:

$$Q_e = \frac{(C_o - C_e) \times V}{m}$$

1

$$\%H = \frac{(C_o - C_e) \times 100\%}{C_o}$$

2

where C_o (mg/L) and C_e (mg/L) are initial and final concentrations of Ni(II) before and after the adsorption process; V (L) and m (g) are the volume of Ni(II) solution and the dosage of RM/CS, respectively.

3. Results And Discussion

3.1. Physicochemical characteristics of RM/CS

The BET surface area of RM and RM/CS are shown in Figures 1a **and b**, respectively. Results inform that the modification significantly enhances the surface area of RM with chitosan. RM, in particular, has a surface area of 68.6 m²/g. The surface area of the modified material reaches 105.7 m²/g after chitosan modification. Clearly, chitosan molecules act as frames to support the structure of the red mud, resulting in increasing the porosity of the material. Consequently, this finding shows that chitosan helps increase the surface area of RM, therefore increasing its adsorption effectiveness. Furthermore, the adsorption-desorption curves in these figures indicate that both materials are mesoporous and suitable for adsorption (Zhu et al. 2017).

Figures 1c **and d** show SEM images of RM and synthesized RM/CS, respectively. RM has a flat and smooth surface, and the particles bind together to form a block, as seen in this diagram. On the contrary, RM/CS shows a porous and rough surface. These results are also consistent with BET analysis, which suggests that the synthesized RM/CS material is convenient and beneficial for the adsorption of heavy metals.

3.2. Effects of some factors on the Ni(II) adsorption

3.2.1. Effect of pH

The point of zero charge (pH_{PZC}) is a significant value to depict the dependence of the pH values on the charge on the surface of the material. At pH values less than pH_{PZC} , the material's surface will be positively charged and vice versa for pH values above the pH_{PZC} (Zhang et al. 2016). Observation in Figure 2a, pH_{PZC} is found at pH of 6.1. The research work of Fei Lyu et al. reported that the pH_{PZC} for red mud modified by colloidal silica and sodium hydroxide was 7.59 (Lyu et al. 2021). In contrast, Tianxue Yang et al. reported the pH_{PZC} of red mud and heat-modified red mud at 900 °C to be 9.8 and 9.5, respectively (Yang et al. 2020).

Since it is an essential factor affecting the removal of heavy metal ions, the effect of pH solution on the Ni(II) removal of RM/CS material was examined at different initial pH levels varying from 2 to 6. From Figure 2b, RM/CS witness a dramatic increase in the adsorption capacity from pH = 2 to pH = 6. This behavior of material can be explained using pH_{PZC} . Because of interactions between functional groups and protons at low pH, the material's surface exhibits positive charges in the pH range of our study.²⁵ As a result, Ni(II) ions compete with protons on the material surface at low pH levels during the adsorption process, resulting in a considerable loss in adsorption efficiency.

On the other hand, there is strong repulsive interaction among the cations in this case due to a large number of positively charged sites on the material's surface. As a result, the Ni(II) adsorption on the material's surface has interfered, and a low adsorption capacity is achieved. The number of negatively charged sites rises by further increasing pH value, thus increasing the electrostatic interactions between Ni(II) ions and negatively charged sites of the material's surface. This leads to the improvement of removal efficiency (Khajavian et al. 2019). In this study, the Ni(II) adsorption efficiency reached the highest value at pH = 6, so it was chosen as the optimal pH for the Ni(II) adsorption of RM/CS.

3.2.2. Effect of contact time

The effect of adsorption time was investigated by varying the contact time between 5 and 240 min. As shown in Figure 2c, the Ni(II) removal efficiency increases as a function of time and reach maximum efficiency at around 180 min. However, the adsorption rate is quick at the beginning, and it slows down after 20 min, which may be ascribed to the fact that there are plenty of adsorption sites on the material's surface in the early stages. Then these adsorption sites gradually decrease during the adsorption process, resulting in the decreasing uptake rate. It is noteworthy that the Ni(II) adsorption obtained the equilibrium after 180 min. Thus, 180 min was chosen as the optimal adsorption time.

3.2.3. Effect of dosage of adsorbent

Figure 2d presents the effect of RM/CS material on the Ni(II) adsorption capacity. As the weight of the material rises from 0.05 g to 0.25 g, the adsorption efficiency gradually rises from 36.21–81.33%. This result can be interpreted by a large number of adsorption sites that are present in the system. Accordingly, the increasing adsorbent amount would lead to the enhancement of adsorption sites, whereas the Ni(II) adsorbate concentration remains constant. Consequently, the uptake capacity and the removal efficiency of Ni(II) are enhanced.

3.3. Adsorption kinetics

In order to investigate the Ni(II) adsorption kinetics of RM/CS, the experimental data were fitted with three kinetic models, including pseudo-first-order, pseudo-second-order, and intra-diffusion kinetic models. Figure 3a describes the Ni(II) adsorption capacity of RM/CS after varying contact time, and the curves are suitable for three of these kinetic models. At the same time, **Table S1** presents the calculated kinematic parameters using a nonlinear method. The results reveal the high agreement of the intra-diffusion kinetic model with the experimental data via the correlation coefficient R^2 , RMSE, and χ^2 error function values are 0.9582, 0.3877, and 0.1752, respectively. Owing to the excellent fit of the intra-diffusion kinetic model to the experimental data and the non-zero value of C, it is expected that the Ni(II) adsorption of RM/CS occurs via different mechanisms, such as electrostatic attraction, chemical interactions, intra-diffusion (Dinh et al. 2020b).

The mass transfer of Ni(II) ions is accurately described via the relationship graph between Q_e and the square root of time $t^{1/2}$ as presented in Figure 3b. Accordingly, the process of eliminating Ni(II) from aqueous solution by RM/CS consists of three stages: (i) First, Ni(II) ions relocate quickly to the surface of the adsorbent (also called external diffusion); (ii) Next, Ni(II) ions migrate from the surface of the adsorbent to the pores of RM/CS (intra-diffusion process); (iii) Finally, the interaction occurs between Ni(II) ions and adsorption sites on the surface of the material by physical or chemical interactions when the adsorption obtains the equilibrium.

During stage 1, Ni(II) ions, positively charged, interact with the material's surface via electrostatic interaction due to their opposite charges. Clearly, at the investigated pH value, functional groups of RM/CS, such as -OH, -COOH, are deprotonated, leading to the surface of RM/CS being charged negatively as the Eqs. 3 and 4. As a result, electrostatic attraction is formed between Ni(II) and RM/CS as the Eqs. 5 and 6.

During stage 2, Ni(II) ions can be diffused into the pores of RM/CS. From the obtained SEM image of RM/CS, it is clear that this material includes many defects, monovacancies, or vacancy clusters, which are suitable for the Ni(II) uptake. This is consistent with the C value calculated from the intra-diffusion model.

At the equilibrium stage, Ni(II) ions continuously interact with the surface of RM/CS via electrostatic attraction and/or the formation of the complex. Notwithstanding, the calculated enthalpy standard (ΔH^θ)

to be smaller than 40 kJ/mol showed that physical adsorption, namely electrostatic interaction, plays a primary role in the Ni(II) removal of RM/CS.

3.4. Adsorption isotherms

Figures 4a, b, and c denote the curves of the Langmuir, Freundlich, and Sips isotherm models of the Ni(II) adsorption of RM/CS at 298K, 313K, and 323K. The corresponding calculated parameters are listed in **Table S2**. As shown in the results, the Sips model is the best fitting model for the experimental data because of its largest correlation coefficient R^2 as well as the smallest RMSE and χ^2 values among three examined models. Notably, the a_s calculated from the Sips model at 298K is zero, which confirms this reduces to the Freundlich model related to the multi-layer adsorption of Ni(II). However, these a_s values at higher temperatures (313K and 323K) differing from zero indicate that the Ni(II) uptake of RM/CS from aqueous solution is mainly based on both monolayer and multi-layer adsorption depended on the Ni(II) initial concentration. This can be explained that the Van der Waals force between Ni(II) ions is decreased when increasing the temperature, resulting in a change from multi-layer adsorption (Freundlich model) to monolayer adsorption (Langmuir model). These findings are absolutely consistent with the evaluated nonlinear parameters at 313K and 323K. For example, comparing the R^2 , RMSE, and χ^2 values show that the Freundlich model gives the best fit with experimental data at 298K and 313K, but these data fit Langmuir more than the former at 323K. Furthermore, the determined Langmuir constant K_L is 0.4992 at 323K ($0 < K_L < 1$), indicating that the Ni(II) uptake of this material is favorable (Al-Ghouti & Da'ana 2020). The maximum adsorption capacity of RM/CS for Ni(II) calculated from the Langmuir model is 31.66 mg/g at 323K, higher than RM and some other natural materials (Table 1).

Table 1
The maximum adsorption capacities of some natural materials

Biosorbents	Optimal adsorption conditions	Q_m (mg/g)	Ref.
Peat	pH = 5, t = 60 min, 298K	61.27	(Bartczak et al. 2018)
Barley straw	pH = 4.85, t = 24 h, 296K	35.80	(Thevannan et al. 2010)
Cashew nutshell	pH = 5, t = 30 min, 303K	18.87	(Kumar et al. 2011)
Tea factory waste	pH = 4, 298K	18.42	(Malkoc &Nuhoglu 2005)
Na - bentonite	pH = 9, 298K	13.97	(Liu &Zhou 2010)
Modified orange peel	pH = 5.5, t = 150 min, 303K	9.82	(Feng et al. 2011)
Coir pith	pH = 7.7, t = 10 min, 303K	9.50	(Ewecharoen et al. 2008)
Rice husk	pH = 6, t = 150 min, 305K	5.52	(Krishnani et al. 2008)
Groundnut shells	pH = 5, t = 120 min, 308K	3.80	(Shukla &Pai 2005)
Green tea waste	pH = 7, t = 180 min, 306K	0.31	(Yang et al. 2016)
Red mud	pH = 5, t = 240 min, 303K	13.69	(Hannachi et al. 2009)
RM/CS	pH = 6, t = 180 min, 323K	31.66	This study

3.5. Thermodynamic studies

The combination of thermodynamic parameters (enthalpy standard (ΔH^0), entropy standard (ΔS^0), and Gibbs free-energy standard (ΔG^0)) and isotherm and kinetic parameters give a complete insight into the behavior of reactions as well as propose technique measures for the treatment of contaminants via the adsorption process (Al-Ghouti &Al-Absi 2020). The relation of these thermodynamic parameters are determined via the Van't Hoff equation as given (Dinh et al. 2020b, Mondal et al. 2017):

$$\Delta G^0 = \Delta H^0 - T\Delta S^0, (7)$$

where ΔG^0 can be determined as the following equation:

$$\Delta G^0 = -RT \ln K_C, (8)$$

where the equilibrium constant of K_C is related to the Langmuir constant (K_L) via the following equation (Dinh et al. 2020a, Zhou &Zhou 2014):

$$K_C = K_L \cdot M \cdot 55.5 \cdot 10^3, (9)$$

where M is the molecular weight of the adsorbate.

By combining equations (7) and (8), the related equation of K_C , ΔH^o and ΔS^o is depicted as:

$$\ln K_C = \frac{\Delta S^0}{R} - \frac{\Delta H^0}{RT}.$$

10

Figure 4d presents the plot of $\ln K_C$ versus $1/T$, and Table 2 lists thermodynamic parameters calculated based on the slope as well as the interception of the graph. Results show that the Ni(II) removal of RM/CS is favorable and spontaneous because the obtained ΔG^o values are negative at all investigated temperatures. This finding has corresponded with the values of n calculated from the Freundlich model. Furthermore, the positive entropy standard ($\Delta S^o = 96.49$ J/mol) shows that during Ni(II) adsorption, the disorderliness in the solid-solution system increases due to a decrease in intermolecular forces temperature rises. This also reconfirms the correlation between isotherm models and examined temperatures in the section of adsorption isotherms. Besides, the negative enthalpy change ($\Delta H^o = -7.27$ kJ/mol) proposes that the Ni(II) uptake is an exothermic process and follows physical adsorption because the absolute value of enthalpy is smaller than 40 kJ/mol (Bayramoglu et al. 2009).

Table 2
Thermodynamic parameters

T (K)	K_L	K_C	ΔH (kJ/mol)	ΔS (J/mol)	ΔG (kJ/mol)
298	0.6278	2055731	-7.27	96.49	-36.02
313	0.5520	1807524			-37.47
323	0.49920	1634630			-38.44

4. Conclusion

RM/CS material was utilized as a novel adsorbent to efficiently remove Ni(II) from aqueous solution. The optimal conditions of uptake were obtained at pH=6 and contact time = 180 min. The intra-diffusion model was the most suitable model to depict the Ni(II) removal from the kinetic and isotherm studies. This metal's adsorption, on the other hand, matched the Sips model. Exothermic and physisorption of Ni(II) on RM/CS were validated by thermodynamic studies. The maximum adsorption uptake calculated from the Langmuir model was 31.66 mg/g at 323K, higher than several other biosorbents. This material performs excellent properties that can be used as a potential adsorbent to remove Ni(II) from aqueous solution.

Declarations

Ethical Approval. Not applicable

Consent to Participate. Not applicable

Consent to Publish. Not applicable

Authors Contributions. Van-Phuc Dinh and H.A. Tuan Kiet designed and directed the research. Van-Phuc Dinh, Thi-Thuy Luu, Thien-Hoang Ho, Duy-Khoi Nguyen and Tu Thi Phuong Nguyen conceived and planned the experiments. Van-Phuc Dinh, Thi-Thuy Luu, Thien-Hoang Ho, Duy-Khoi Nguyen, Tu Thi Phuong Nguyen, H.A. Tuan Kiet carried out the experiments, analysed the data, and performed the isotherm and kinetic model analyses. Van-Phuc Dinh, Thi-Thuy Luu and H.A. Tuan Kiet wrote the manuscript with inputs from all authors. All authors discussed the results and contributed to the final manuscript. All authors read and approved the final manuscript.

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Availability of data and materials. Not applicable

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Figures

Figure 1

BET surface area of RM (a) and RM/CS (b); SEM images of RM (c) and RM/CS (d)

Figure 2

pHPZC of RM/CS (a); Effects of pH (b), adsorption time (c), and adsorbent dosage (d) on the Ni(II) uptake of RM/CS

Figure 3

Plots of kinetic models of the Ni(II) adsorption of RM/CS (a) and Q_e vs $t_{1/2}$ (b)

Figure 4

Plots of isotherm models of the Ni(II) adsorption of RM/CS at 298K (a), 313K (b), and 323K (c); $\ln K_c$ vs $1/T$ (d)

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- [Picture1.png](#)
- [SUPPLEMENTARYINFORMATION.docx](#)