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Removal of vanadate anion by calcined Mg/Al–CO₃ layered double hydroxide in aqueous solution

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Abstract: Mg/Al–CO₃ layered double hydroxide (LDH2) with Mg(II):Al(III) molar ratio of 2:1 was synthesized by co-precipitation method and its calcined product Mg₂Al–CLDH(CLDH2) was prepared by heating Mg₂Al–LDH at 773 K for 6 h. Removal of vanadate anion (VO_4^{3-}) from aqueous solution on CLDH2 was studied. Batch studies were carried out to address various experimental parameters such as Mg/Al molar ratio, adsorbent dosage, initial concentration of solution, contact time and temperature. Vanadate was removed effectively at the optimized experimental conditions. The adsorption kinetics data fitted the pseudo-first-order model. Isotherms for adsorption vanadate by CLDH2 at different solution temperatures were well described using the Langmuir and Freundlich equations, and the isotherm parameters were calculated using linear regression analysis. The adsorption data fitted the langmuir model with good values of the correlation coefficient (R^2 >0.999). The negative value of ΔG^{Θ} and the positive value of ΔH^{Θ} indicate that the adsorption processes are spontaneous endothermic in nature. The mechanism of adsorption suggests that the surface adsorption is the main process.

Key words: layered double hydroxide; vanadate; adsorption; kinetics; thermodynamics

1 Introduction

Layered double hydroxides (LDHs), known as synthetic anionic clays or hydrotalcite-like compounds (HTICs), are an important class of ionic lamellar solids. LDHs have the general formula $[M_{1-x}^{II}M_x^{III}(OH)_2]^{x+}$. $(A^{n-})_{x/n}\cdot mH_2O$, where M^{II} is a divalent metal cation $(Mg^{2+}, Zn^{2+}, Cu^{2+}, etc)$; M^{III} is a trivalent (Al³⁺, Cr³⁺, Fe³⁺, etc) cations that occupies octahedral sites in the hydroxide layers; A^{n-} is an exchangeable interlayer anion; *x* is defined as the $M^{III}/(M^{II}+M^{III})$ mole ratio that determines layer charge density [1,2].

Recently, LDHs have attracted considerable attention for their potential applications such as catalysts, catalyst supports, ion exchangers and adsorbents [3–7]. By heating LDH at 500 °C, the interlayer CO_3^{2-} can be removed and the calcination of LDH transforms them into mixed oxides (CLDH) which will return to their original structure through rehydration and incorporation of anions in aqueous medium. This phenomenon is known as "memory effect", and the processes are expressed by Eqs. (1) and (2). Therefore, LDH adsorption efficiency can be significantly increased by the calcination process.

Due to their memory effect property, CLDH is suitable for adsorption of anionic species.

$$[M_{1-x}^{II}M_{x}^{III}(OH)_{2}](CO_{3})_{x/2} \rightarrow M_{1-x}^{II}M_{x}^{III}O_{1+x/2} + x/2CO_{2} + H_{2}O \qquad (1)$$

$$M_{1-x}^{II}M_{x}^{III}O_{1+x/2} + (x/n)A^{n-} + [1 + (x/2) + m]H_{2}O \rightarrow xOH^{-} + [M_{1}^{II}M_{x}^{III}(OH)_{2}](A^{n-})_{x/n} \cdot mH_{2}O \qquad (2)$$

Vanadate, a water soluble salt, is widely used in chemical production, industry, machinery steel manufacturing, automobile and catalyzer [8,9]. But for most producers, the waste water from the production is seldom treated. Its direct release to water is an environmental concern, resulting in deterioration of water quality. The sewage containing vanadium compounds does serious harm to human health [10-12]. So, there is an increasing demand for the removal of vanadate from polluted water. Various techniques such as ion exchange, adsorption, chemical precipitation and biological treatment have been used for vanadate removal [13–15]. The adsorption method proves to be more effective due to its lower cost, higher uptake capacity, faster regeneration kinetics and easier operation.

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From the survey of the literatures, no information for vanadate removal in detail by CLDH was available. Therefore, it is important to research the adsorption of vanadate by CLDH. In this work, LDH was synthesized, calcined and then used for removal of vanadate from water. The effects of several conditions on the removal process including Mg/Al molar ratio, adsorbent dosage, initial concentration of solution, contact time and temperature have been investigated. The equilibrium isotherm and kinetic model for the adsorption process were also studied to evaluate the viability and effectiveness of the process. The adsorption mechanism was also discussed with the supports of characterization results.

2 Experimental

2.1 Reagents

 $Mg(NO_3)_2 \cdot 6H_2O$ and $Al(NO_3)_3 \cdot 9H_2O$ were obtained from Bodi Chem. Co. Ltd. (Tianjing, China). NaOH, Na₂CO₃, HCl and C₂H₅OH (95%) were purchased from Taishan Chem. Co. Ltd (Guangzhou, China). Vanadate was supplied by National Drug Group Co. Ltd. (Beijing, China). Deionized water was prepared by twice distillation.

2.2 Preparation of LDH and CLDH

Mg/Al-LDH with Mg (II) to Al (III) molar ratio of 2:1 was prepared by the co-precipitation method [16]. In this method, solution A containing 0.08 mol of $Mg(NO_3)_2 \cdot 6H_2O$ and 0.04 mol of $Al(NO_3)_3 \cdot 9H_2O$ in 150 mL of deionised water was slowly added to a vigorously stirred solution B containing 0.12 mol of NaOH and 0.12 mol of Na₂CO₃ in 150 mL deionised water. The solution pH was maintained slightly higher than 10. The precipitate was aged at 80 °C for 12 h in a thermostatic bath. It was then centrifuged, washed thoroughly with deionised water and C2H5OH till neutral pH and dried at 80 °C overnight. Two more Mg/Al-LDH samples with Mg/Al molar ratio of 3:1(Mg₃Al) and 4:1(Mg₄Al) were also prepared according to the similar method. The CLDH was obtained by calcining the LDH in a muffle furnace at 500 °C for 6 h. The LDH and calcined product were stored in a desiccator prior to adsorption studies.

2.3 Adsorption experiments

The solution of vanadate for adsorption studies was prepared from analytical reagent (AR) grade Na_3VO_4 ·12H₂O. Adsorption of vanadate on CLDH was carried out in a 250 mL stoppered conical flask with variation of solution concentration (50–500 mg/L), adsorbent dose (0.4–4.0 g/L), and temperature (20–40 °C). The solution along with prearranged amount of CLDH was shaken immediately after mixing by SHY-2 water shaker bath for a predetermined temperature and contact time. After reaction, the adsorbent conserved for latter detection was separated from the mixture by filtration, and the residual vanadate in the filtrate was determined by Optima 5300 DV ICP-OES (Perkin Elmer Instruments, America).

The adsorption capacity Q_e and the removal rate R of vanadate are calculated from the following equations:

$$Q_{\rm e} = \frac{(C_0 - C_{\rm e})V}{m} \tag{3}$$

$$R = \frac{C_0 - C_e}{C_0} \times 100\%$$
 (4)

where C_0 is the initial concentration of the vanadate in solution (mg/L); C_e is the equilibrium concentration of vanadate in solution (mg/L); Q_e is the equilibrium adsorption capacity (mg/g); *m* is the adsorbent dry mass (g); *V* is the suspension volume (L).

2.4 Analytical methods

The LDH (before and after adsorbing) and CLDH (before and after adsorbing) were characterized by X-ray diffraction (XRD) and scanning electron microscopy (SEM). Powder X-ray diffraction patterns were recorded using a Rigaku Rint 6000 powder X-ray diffractometer with Ni-filtered Cu K_{α} (λ =1.5406 Å) radiation at voltage of 40 kV and current of 250 mA. The sample was scanned in steps of 0.02° (2 θ) in the range from 5–70° with a count time of 4 s per step. Scanning electron microscopy (SEM) images were obtained on a JEOL 5190 microscope operating at an acceleration voltage of 25 kV.

3 Results and discussion

3.1 Effect of Mg/Al molar ratio

In order to find out the effect of Mg/Al molar ratio on the removal of vanadate, Mg/Al–LDH and CLDH with three different ratios such as 2:1, 3:1 and 4:1 were subjected to vanadate adsorption in the same condition. It was observed from Fig. 1 that the removal rate decreased with increasing Mg/Al molar ratio. This could be due to the fact that Mg₂Al contains a higher amount of Al³⁺ and the net positive charge on the hydroxide layer was higher than Mg₃Al and Mg₄Al. It was also found that adsorption of vanadate on CLDH was appreciably higher than the LDH. Therefore, Mg₂Al–CLDH (CLDH2) was used for adsorbent in subsequent adsorption experiments.

3.2 Crystalline structure

The XRD patterns of $Mg_2Al-LDH$ (LDH2) before and after vanadate adsorbing, and $Mg_2Al-CLDH$



Fig. 1 Effect of Mg-Al molar ratio on adsorption of vanadate

(CLDH2) before and after vanadate adsorbing are shown in Fig. 2. The XRD pattern of LDH2 is shown in Fig. 2(a). It is found that more intense peaks are located at lower 2θ while less intense lines are present at higher 2θ values. The sharpness of the (003) (0.7657 nm) and (006)(0.3803 nm) peaks demonstrates the high degree of crystallinity and layered structure of the sample, which agrees well with the previous research [17]. Figure 2(b) shows the pattern of LDH2 after vanadate adsorbing, the features at (003) (0.7662 nm) and (006) (0.3805 nm) are essentially retained after vanadate adsorbing, which indicates that the adsorption of vanadate on LDH2 is using surface adsorption. Figure 2(c) shows the decomposition of LDH2 by calcined at 773 K for 6 h. After calcination, the (003) and (006) reflections practically disappears, indicating that the LDH2 structure is destroyed and the stacking of the lays becomes disordered. Only few diffused peaks at 62.59° and 43.21° are observed which are attributed to MgAl(O). Figure 2(d) shows the diffraction patterns of CLDH2 after vanadate adsorbing. The reappearance of characteristic



Fig. 2 XRD patterns of $Mg_2Al-LDH$ before (a) and after (b) vanadate adsorbing, $Mg_2Al-CLDH$ before (c) and after (d) vanadate adsorbing

layered structure of LDH2 with (003) (0.7649 nm) and (006) (0.3817 nm) indicates that the adsorption of vanadate and carbonates anions is using the "memory effect" of CLDH2.

3.3 Morphological structure

The SEM images of LDH2 and CLDH2 before and after vanadate adsorbing are obtained and presented in Fig. 3. The microstructure of the LDH2 can be clearly observed from Figs. 3(a) and (b), which show aggregated mass of irregularly shaped tiny particles with a highly porous structure. After calcination, the adsorbent becomes into irregular particles which disperse well and the particle size distributes from a few nano to microns as shown in Figs. 3(c) and (d). The small particle size and the porous structure provide a good adsorption capacity for vanadate. As seen from Figs. 3(e) and (f), the reconstructed CLDH2 surface with a petal-like morphology is evident, which is different from that of the clean CLDH2. Compared with the morphology of LDH2 platelets, the full extent of adsorption of vanadate onto CLDH2 can't be observed, which may be due to the high level of crystallinity of this adsorbent.

3.4 Effect of adsorbent quantity

The effect of dosage of adsorbent on the removal of vanadate at temperature of 30 °C is investigated with the initial concentration of 170 mg/L and the results are shown in Fig. 4. As seen from Fig. 4, the removal rate is increased with increasing amount of adsorbents until 2 g/L which is owing to the increasing presence of adsorption sites. Further increase in the dosage of adsorbent has no positive effect on the removal rate. But the adsorption capacities gradually decrease. This may be explained by limitation of the vanadate concentration. At higher adsorbent dose, the ratio of initial number of moles of vanadate to the CLDH2 is low and the adsorbed exchanged sites cannot reach saturation point. Therefore, considering both of the removal efficiency and the sufficient use of the adsorbent, adsorbent dose of 2 g/L CLDH2 is used in subsequent adsorption experiments.

3.5 Effect of initial vanadate concentration and contact time

In order to find out the effect of contact time at various initial concentrations (50–300 mg/L) on vanadate removal by CLDH2, experiments were carried out at ambient temperature (30 °C) for contact period up to 12 h. The results are depicted in Fig. 5. It indicates the removal rate increases with increasing contact time and the equilibrium time increases with the increase of initial vanadate concentration. At low initial concentrations, it can achieve adsorption balance for 1-2 h, and vanadate is almost removed completely from the aqueous solution



Fig. 3 SEM images of Mg₂Al-LDH (a) and (b), Mg₂Al-CLDH (c) and (d), and Mg₂Al-CLDH (e) and (f) after vanadate adsorbing



Fig. 4 Effect of adsorbent dosage on vanadate removal by $\ensuremath{\mathsf{CLDH2}}$



Fig. 5 Effect of contact time at various initial concentrations on vanadate removal by CLDH2

in a short time, indicating that the overall adsorption process is based mainly on external surface adsorption. The removal rate of vanadate begins to decline at high concentration, owing to the limitation of adsorbent dose. Clearly, the equilibrium adsorption capacity of vanadate increases with the increase of initial vanadate concentration. At higher concentrations, the higher equilibrium adsorption capacity increases due to an increased ratio of initial number of moles of vanadate to the available surface areas.

3.6 Adsorption kinetics

The adsorption kinetics is an important aspect of the pollutant removal process control. The adsorption of the studied metal ions onto CLDH2 as a function of contact time is investigated. Since temperature is a highly significant parameter in the adsorption process, the kinetics for removal of vanadate at different temperatures is studied. Lagergren's pseudo-first-order model, Ho's pseudo-second-order model and Chien's Elovich model are the most widely used models in description solid, liquid adsorption kinetics to predict the involved mechanism in the adsorption process. In this work, kinetic analyses are made at various temperatures depending on contact time by these three equations.

The pseudo-first-order model is described by the following equation:

$$\frac{\mathrm{d}Q}{\mathrm{d}t} = k_1(Q_\mathrm{e} - Q_t) \tag{5}$$

where Q_t and Q_e (mg/g) are the amounts of adsorbed vanadate at time *t* and at equilibrium, respectively; k_1 (min⁻¹) is the first-order rate constant.

Equation (5) can be expressed by the following linearization equation:

$$\ln(Q_{\rm e} - Q_t) = \ln Q_{\rm e} - k_1 t \tag{6}$$

According to Ho and McKay model, the differential equation for pseudo-second-order model is

$$\frac{\mathrm{d}Q_t}{\mathrm{d}t} = k_2 (Q_\mathrm{e} - Q_t)^2 \tag{7}$$

where Q_t and Q_e are the amounts of adsorbed vanadate at time *t* and at equilibrium, respectively; $k_2 (\min^{-1}(\text{mg/g})^{-1})$ is the pseudo-second-order rate constant.

The linearization of Eq. (7) is given as follows:

$$\frac{t}{Q_t} = \frac{1}{k_2 Q_e^2} + \frac{t}{Q_e}$$
(8)

The kinetics of adsorption of vanadate onto CLDH2 is also examined using Elovich equation:

$$Q_t = \frac{1}{\beta} \ln \alpha \beta + \frac{1}{\beta} \ln t \tag{9}$$

where α is the initial adsorption rate (mg/min); β is the desorption constant (g/mg); Q_t is the adsorption capacity at time *t*.

The adsorption data obtained by experiments have been analyzed by the linear forms of pseudo-first-order model, pseudo-second-order model and Chien's Elovich model, as shown in Figs. 6–8, respectively. The obtained constants and parameters for each model are listed in Table 1. From Figs. 6–8, it is found that they are all applicable to fitting the experimental data. However, the pseudo first-order kinetic is the most appropriate as the the correlation coefficient R^2 value is the highest. This indicates that the adsorption complies with the pseudo-first-order reaction.



Fig. 6 Pseudo-first-order plots for vanadate adsorption on CLDH2



Fig. 7 Pseudo-second-order plots for vanadate adsorption on CLDH2

3.7 Adsorption isotherm studies

The adsorption isotherm provides a relationship between the concentration of metal anion in an adsorption system and the amount of metal anion adsorbed onto the solid phase when the two phases are at a fixed temperature at equilibrium. For the liquid–solid system, adsorption isotherm equations usually use the Hai-lei SONG, et al/Trans. Nonferrous Met. Soc. China 23(2013) 3337-3345



Fig. 8 Elovich plots for vanadate adsorption on CLDH2

Langmuir equilibrium model and the Freundlich equilibrium model.

The Langmuir isotherm model is described by the following equation:

$$Q_{\rm e} = \frac{K_{\rm L} Q_{\rm max} C_{\rm e}}{1 + K_{\rm L} C_{\rm e}} \tag{10}$$

where Q_e (mg/g) is the amount of VO₄³⁻ removed per gram of adsorbent; Q_{max} (mg/g) is the maximum adsorption capacity; C_e (mg/L) is the equilibrium VO₄³⁻ concentration; K_L (L/mg) is the Langmuir constant related to adsorption energy.

The linear form of Langmuir equation can be expressed as follows:

$$\frac{C_{\rm e}}{Q_{\rm e}} = \frac{1}{K_{\rm L}Q_{\rm max}} + \frac{C_{\rm e}}{Q_{\rm max}}$$
(11)

The Freundlich model is expressed by the following equation:

$$Q_{\rm e} = K_{\rm F} C_{\rm e}^{1/n} \tag{12}$$

where Q_e (mg/g) is the equilibrium concentration of solute per gram of adsorbent; C_e (mg/L) is the equilibrium aqueous concentration of the solute; K_F and *n* are Freundlich constants which are related to adsorption and intensity of adsorption, respectively.

Equation (12) can be linearized in its logarithmic form, which allows the determination of Freundlich constants as follows:

$$\ln Q_{\rm e} = \ln K_{\rm F} + \frac{1}{n} \ln C_{\rm e} \tag{13}$$

The adsorption data obtained for equilibrium conditions have been analyzed by the linear forms of the two kinds of isotherms in Figs. 9 and 10. The values of $K_{\rm L}$, 1/n, $K_{\rm F}$, $Q_{\rm max}$ and the correlation coefficients for Langmuir and Freundlich models are given in Table 2. It can be seen from Table 2 that the correlations coefficient R^2 values for Langmuir model are higher than those of Freundlich model. Thus we can deduce that adsorption isotherms for CLDH2 are better represented by Langmuir model, which may be due to the homogenous distribution of active sites on the CLDH2 surface.

The study of the temperature effect on vanadate



Fig. 9 Langmuir plots for vanadate adsorption on CLDH2 at different temperatures



Fig. 10 Freundlich plots for vanadate adsorption on CLDH2 at different temperatures

Table 1 Kinetic parameters for vanadate adsorption by CLDH2

Temperature/K -	Pseudo-first-order constant		Pseudo-second-order constant		Elovich equation		
	k_1/\min^{-1}	R^2	$k_2/(g \cdot mg^{-1} \cdot min^{-1})$	R^2	$\alpha/(\text{mg·min}^{-1})$	$\beta/(g \cdot mg^{-1})$	R^2
293	0.01195	0.9994	0.000106	0.9797	209.501	0.0383	0.9714
303	0.01396	0.9998	0.000152	0.9903	147.524	0.0446	0.9834
313	0.01708	0.9996	0.000143	0.9966	124.286	0.0513	0.9905

 Table 2
 Isotherm
 parameters
 for
 vanadate
 adsorption
 on

 CLDH2

Temperature/	Langmuir		Freundlich		
К	$K_{\rm L}/({\rm L}\cdot{\rm g}^{-1})$	R^2	$K_{\rm F}/({\rm L}\cdot{\rm g}^{-1})$	n	R^2
293	0.352	0.9995	58.90	15.02	0.9599
303	0.319	0.9992	64.39	13.77	0.9897
313	0.337	0.9991	71.50	14.66	0.9989

adsorption on CLDH2 enable us to determine the thermodynamic parameters (ΔG^{Θ} , ΔH^{Θ} and ΔS^{Θ}) of these reactions by the following equation:

$$\ln K_{\rm d} = \frac{\Delta S^{\Theta}}{R} - \frac{\Delta H^{\Theta}}{RT}$$
(14)

$$\Delta G^{\Theta} = \Delta H^{\Theta} - T \Delta S^{\Theta} \tag{15}$$

where *R* is molar gas constant; *T* is the thermodynamic temperature, K; K_d is the distribution coefficient, which is calculated with the following equation:

$$K_{\rm d} = \frac{Q_{\rm e}}{C_{\rm e}} \tag{16}$$

The thermodynamic parameters calculated from the equations are reported in Table 3. The positive values of ΔS^{Θ} suggested VO₄³⁻ ions are not too restricted in the beads and the increasing randomness at the solid/solution interface during the adsorption. The positive values of ΔH^{Θ} indicated that adsorption of vanadate on CLDH2 is an endothermic nature process. The ΔG^{Θ} values decrease from -1.053 to -3.286 kJ/mol when the temperature increases from 293 to 313 K, suggesting the more adsorbable of vanadate with increasing temperature. Furthermore, the negative values of ΔG^{Θ} at different temperatures indicated the feasible and spontaneous nature of the adsorption of vanadate onto CLDH2.

 Table 3 Gibbs free energy, enthalpy and entropy changes for vanadate adsorption on CLDH2

Temperature/K	ΔG^{Θ} / (kJ·mol ⁻¹)	$\Delta H^{\Theta}/$ (kJ·mol ⁻¹)	ΔS^{Θ} / (J·mol ⁻¹ ·K ⁻¹)
293	-1.043		
303	-2.169	31.67	111.63
313	-3.286		

3.8 Mechanism study

In order to further study the adsorption process, the XRD patterns of Mg_2Al -CLDH after adsorption of vanadate at different initial concentrations are shown in Fig. 11. Figure 11 shows that the reduction in peak intensity and the increase in peak width after adsorption of vanadate indicate a decrease in crystallization from the overall look. At low concentrations no change in the

interlayer spacing as measured by XRD (Fig. 11(b)) indicates mainly surface adsorption and no structural change in the matrix, that is to say, VO_4^{3-} anions does not intercalate between the interlayer. At high concentrations, the corresponding CLDH2 exhibits new reflections corresponding to phases with increased basal spacing, which indicates that part of VO_4^{3-} ions are intercalated in the hydrotalcite interlayer. Similar results have been reported for the adsorption of inorganic anions in CLDH2 [18]. It shows that at high concentrations the adsorption is not only the surface adsorption, but also reconstruction adsorption and ion-exchange. In short, no matter concentrations are high or low, surface adsorption dominates the adsorption process.



Fig. 11 XRD patterns of CLDH2 after adsorption of vanadate at various initial concentrations: (a) 0 mg/L; (b) 50 mg/L; (c) 100 mg/L; (d) 200 mg/L; (e) 300 mg/L; (f) 500 mg/L

With the supports of the above analysis, the mechanism of the adsorption process is discussed as follows.

CLDH2 is obtained by calcining LDH2, the process of calcination can be expressed as Eq. (17). CLDH2 uptake anions from solution mainly have three mechanisms: first, external surface adsorption; second, intercalation by reconstruction of the LDH2 structure; third, anion exchange. Making a comprehensive view of the results and discussions above, the adsorption process of vanadate onto the CLDH2 may be separated into three discriminating phases. In the first phase, the surface adsorption occur rapidly within 60 min, indicating a physic adsorption on the external surface of CLDH2 dominates the adsorption process. In the second phase, intercalation of vanadate anion and carbonate anion reconstruct the LDH2 structure. The process of reconstruction can be expressed as Eqs. (18) and (19).

$$[Mg_2Al(OH)_8]_2CO_3 \cdot xH_2O \longrightarrow$$
$$Mg_4Al_2O_9 + CO_2 + (x+8)H_2O$$
(17)

$$Mg_4Al_2O_9 + 8H_2O + CO_2 \rightarrow [Mg_2Al(OH)_8]_2CO_3 (18)$$

 $3Mg_4Al_2O_9 + 27H_2O + 2VO_4^{3-} \rightarrow$

 $2[Mg_2Al(OH)_8]_3(VO_4) + 6OH^-$ (19)

According to Eq. (19), OH⁻ is released during the reconstruction process. Figure 11 shows the basal widening of Mg₂Al–LDH after reconstruction, which indicates that vanadate anions are probably intercalated between the interlayer. In the third phase, the slow and trifling interlayer or layer ion exchange between VO₄³⁻ and CO_3^{2-} on the LDH2 layers are happened as

$$3[Mg_2Al(OH)_8]_2CO_3 + 2VO_4^{3-} \rightarrow 2[Mg_2Al(OH)_8]_3(VO_4) + 3CO_3^{2-}$$
 (20)

The CLDH2 after adsorption can be reused by calcination at 500 °C with the adsorbed vanadate being almost completely removed. It is found that the removal rate of vanadate by the regenerated CLDH2 declined by 10% compared with the original use, which may be due to the decreasing crystallinity during regeneration process. Considering the small reducing of adsorption capacity, this material can be recycled for further use and has great potential for waste water treatment.

4 Conclusions

1) CLDH2 can be used as an effective adsorbent for the removal of vanadate from aqueous solution. It is found that vanadate can be removed effectively by optimizing experimental conditions with a removal rate of 99.9% for 100 mg/L.

2) The kinetic and sorption data fit well with the first order kinetic model and the Langmuir model, respectively, with good values of the determination coefficient. The negative value of ΔG^{Θ} and the positive value of ΔH^{Θ} indicate that the adsorption processes are spontaneous endothermic in nature.

3) The mechanism of adsorption suggests that the surface adsorption is the main process.

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煅烧烧型 Mg/Al-CO₃ 双金属氧化物对 水溶液中钒酸根离子的脱除

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摘 要:采用共沉淀法合成 Mg/Al 摩尔比为 2:1 的层状双金属氢氧化物 Mg/Al-LDH,并在 500 °C 下煅烧 6 h 得 到煅烧双金属氢氧化物 Mg/Al-CLDH。研究 Mg/Al-CLDH 对钒酸根的吸附性能,探讨镁铝摩尔比、吸附剂用量、 溶液初始浓度、时间和温度等因素对吸附效果的影响,并对其吸附过程的动力学和热力学过程进行研究。研究表 明: Mg/Al-CLDH 对钒酸根的吸附效果很好。该吸附过程符合准一级动力学方程,平衡吸附等温线很好地符合 Langmuir 方程,并且 Langmuir 方程拟合常数高达 0.999。通过热力学计算发现 ΔG[®] 为负值, ΔH[®] 为正值,表明 此吸附本质上是吸热自发过程。吸附机理研究表明此吸附主要是表面吸附。

关键词: 层状双金氢氧化物; 钒酸根离子; 吸附; 动力学; 热力学

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