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Keywords: silicate, alumina, hydrocalumite, Adsorption, black dross

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Article

The Removal of Silicate(IV) by Adsorption onto Hydrocalumite from the Sodium Hydroxide Leaching Solution of Black Dross

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Abstract: Alkaline leaching of mechanically activated black dross resulted in an aluminate(III) solution with a small amount of silicate(IV). To obtain pure aluminate(III) solution, the removal of silicate(IV) from the alkaline leaching solution was investigated by adsorption with hydrocalumite ($\text{Ca}_2\text{Al}(\text{OH})_6\text{Cl}\cdot 2\text{H}_2\text{O}$). The hydrocalumite was synthesized by the coprecipitation method. The characterization of the synthesized hydrocalumite was analyzed via X-ray diffraction (XRD), scanning electron microscopy (SEM) images and Fourier-transform infrared spectroscopy (FTIR). In our experimental conditions, silicate(IV) was selectively adsorbed onto hydrocalumite over aluminate(III). The reaction time greatly affected the removal percentage of aluminate(III) owing to mass action effect. When the reaction time was longer than 2 h, no aluminate(III) was adsorbed onto hydrocalumite and thus it was possible to selectively remove silicate(IV). When the dosage of hydrocalumite was in excess, the removal percentage of silicate(IV) was rather reduced. Complete removal of silicate(IV) from the solution was accomplished in the reaction temperature between 50 and 70 °C. By selective adsorption of silicate(IV) from the solution at the optimum condition (30 g/L hydrocalumite, 50 °C, 400 rpm, and 2 h), an aluminate(III) solution with purity higher than 99.9% was obtained. The adsorption of silicate(IV) onto hydrocalumite followed the Freundlich isotherm.

Keywords: black dross; adsorption; hydrocalumite; alumina; silicate

1. Introduction

Aluminum dross contains alumina, metal oxides and nitrides, and some salts [1]. Black dross results from the treatment of used aluminum cans to recover aluminum. Since alumina is amphoteric, alumina can be dissolved in either acidic or alkaline solution. In acid leaching, some oxides in black dross are dissolved together with alumina and thus the removal of these impurities is necessary to obtain pure alumina solution [2]. In contrast, an alkaline leaching solution has some selectivity for alumina because the oxides of Ca, Fe, and Mg would not dissolve in this medium, resulting in pure aluminate(III) solution with a small amount of silicate(IV) [3].

Various methods have been developed to remove silicate(IV), such as adsorption, coagulation, nano-filtration, reverse osmosis, and precipitation [4–6]. In precipitation with CaO, CaO reacts with $\text{Al}(\text{OH})_4^-$ and $\text{SiO}_2(\text{OH})_2^{2-}$ in the solution to form hydrogarnet. This might lead to simultaneous co-precipitation of aluminate(III) and silicate(IV). Among these methods, the adsorption using various adsorbents has been recognized as an effective and potential method. In a previous study, removal of silicate(IV) by reaction with polyacrylamide(PAM) was investigated [7]. The removal percentage of silicate(IV) increased linearly as the PAM dosage increased. However, the viscosity of the slurries increased with an increase of PAM dosage. The high viscosity led to some difficulty in separating

silicate(IV) and in filtration. Therefore, PAM is not desirable to the removal of silicate(IV) when the concentration of silicate(IV) in the solution is high.

Layered double hydroxides (LDHs) are well known as the catalyst carriers and adsorbent materials. They can be represented as the general formula, $[M_{1-x}^{2+}M_x^{3+}(\text{OH})_2]A_{x/n}\cdot y\text{H}_2\text{O}$, where M^{2+} and M^{3+} represent divalent and trivalent metal cations, A and x represent interlayer anions and the concentration ratio defined as $[M^{3+}]/([M^{2+}]+[M^{3+}])$ [8,9]. It has been reported that $\text{Mg}_6\text{Al}_2(\text{OH})_{16}(\text{CO}_3)\cdot 4\text{H}_2\text{O}$ could remove silicate(IV) from aluminate(III) solution but its loading capacity for silicate(IV) is very low, which prevents its application for the removal of silicate(IV) [10]. Hydrocalumite ($\text{Ca}_2\text{Al}(\text{OH})_6\text{Cl}\cdot 2\text{H}_2\text{O}$) is a kind of trigonal crystals with a flat hexagonal structure, in which the chloride ion can be exchanged with either bivalent or trivalent anions.

In this work, hydrocalumite was selected for the removal of silicate(IV) from the NaOH leaching solution of black dross on the basis of the following reasons. Hydrocalumite occurs in the hydrated cement paste and thus can be cheaply prepared. Compared to other adsorbents, hydrocalumite has several advantages such as the ease of synthesis and high adsorption capacity.

In our previous work on the recovery of alumina from black dross, the NaOH leaching of the mechanically activated dross suppressed the dissolution of silica and resulted in a leaching solution where the purity of aluminate(III) was 98% [11]. Although the purity of aluminate(III) in the leaching solution is relatively high, removal of silicate(IV) from the leaching solution would increase the purity of aluminate(III). Therefore, the removal of silicate(IV) by adsorption with hydrocalumite was attempted in this work. For this purpose, the effect of the dosage of hydrocalumite, reaction temperature, reaction time and the concentration of silicate(IV) was investigated. The optimum conditions for the complete removal of silicate(IV) by adsorption with hydrocalumite were obtained and the adsorption isotherm was identified.

2. Materials and Methods

2.1. Materials

The synthetic leach solution of mechanically activated alumina dross was prepared by dissolving AlCl_3 (Daejung Chemicals and Metals Co., Ltd., Shiheung, Korea, >95%) and SiO_2 (Duksan Pure Chem. Co., Ltd., Ansan-si, Korea, 99%) in sodium hydroxide solution (Duksan Pure Chem. Co., Ltd., Ansan-si, Korea). In this work, AlCl_3 was selected on the basis of two main reasons. Firstly, aluminum salts are more soluble than other types of aluminum compounds. Secondly, although the salts in black dross are removed by washing with water at high temperatures [12,13], some of chloride ions exist in the leaching solution of black dross. Therefore, the use of AlCl_3 is better in preparing the synthetic leaching solution. Doubly distilled water was used as the solvent. The concentration of Al(III) and Si(IV) in 5 M NaOH solution was fixed at 0.4815 M and 0.0054 M, respectively.

2.2. Silicate(IV) Removal Experiment

For the removal of silicate(IV), various dosages of hydrocalumite was added to 50 mL of the synthetic leaching solution in covered 100 mL beakers. The experiments were done by employing a heating mantle with a magnetic stirrer bar. The mixture of hydrocalumite and leaching solution was stirred at 400 rpm at desired temperature. The slurry samples were taken at desired time intervals and filtered by using vacuum filtration. The filtered residues were washed several times with distilled water until the filtrate became completely clean and then dried at 60 °C for 48 h. The morphology of the residue after adsorption was investigated by Field Emission scanning electron microscopy (FE-SEM) (S-4800, Hitachi, Tokyo, Japan). The concentration of silicate(IV) and aluminate(III) in the filtrate was measured by inductively coupled plasma optical emission spectrometer (ICP-OES, Spectro Arcos, Cleve, Germany). The removal percentage of silicate(IV) and aluminate(III) (R, %) was calculated by using Equation (1)

$$R = (C_0 - C_f)/C_0 \times 100 \quad (1)$$

where C_0 and C_f represent the concentration of silicate(IV) and aluminate(III) ion in the solution, respectively, before and after the experiment.

2.3. Preparation of Hydrocalumite

Hydrocalumites were prepared by heating 0.5 M CaCl_2 (Duksan Pure Chem. Co., Ltd., Ansan-si, Korea) (300 mL) to the desired temperature (50 °C) and adding 200 mL NaAlO_2 (containing 0.25 M $\text{Al}(\text{OH})_3$ (Duksan Pure Chem. Co., Ltd., Ansan-si, Korea) and 0.5 M NaOH (Duksan Pure Chem. Co., Ltd., Ansan-si, Korea)) using burette, whilst stirring at 300 rpm. The mixed solution was stirred for 1 h. The slurry samples at desired time intervals were taken and filtered by using vacuum filtration. In each case, the precipitates were separated by vacuum filtration, washed several times with warm deionized water until the filtrate became completely clean and then dried at 60 °C for 48 h. The structure of hydrocalumite was characterized by X-ray diffraction (XRD) (D8 Advance (Bruker AXS, Karlsruhe, Germany)) with $\text{Cu K}\alpha$ (40 kV/40 mA, $\gamma = 0.15406$ nm) radiation.

3. Results and Discussion

3.1. Characterization of Synthesized Hydrocalumite

Figure 1 shows the XRD pattern of the synthesized hydrocalumite. The typical XRD peaks of hydrocalumite (e.g., $2\theta = 11.2^\circ, 22.4^\circ, 28.4^\circ, 31.2^\circ, 38.6^\circ$) were identified and agreed well within the standard card profile of the monoclinic LDH structure [14], indicating the successful synthesis of the hydrocalumite crystals.

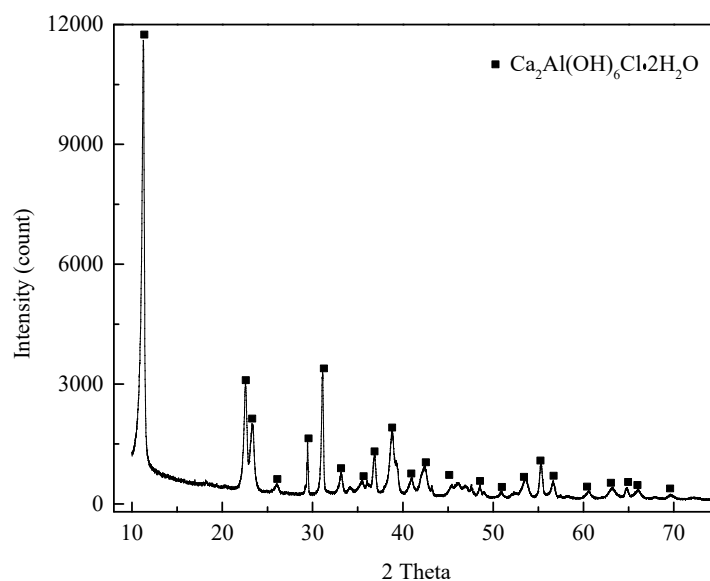


Figure 1. The XRD pattern of the hydrocalumite synthesized in this work.

FT-IR was employed to investigate the characteristic vibrations of the hydrocalumite and is shown in Figure 2. A partial assignment of the peaks is as follows: The features at 524 and 790 cm^{-1} were due to the Al-OH stretching vibration [15]. The peak at 1630 cm^{-1} was attributed to the HO-H vibration of interlayer water. The peaks at 3473 and 3641 cm^{-1} were ascribed to the $-\text{OH}$ groups vibration of structural water. The weak band at 1416 cm^{-1} was the CO_3^{2-} stretching vibration, indicating the possible incorporation of CO_2 during the synthesis process [16,17].

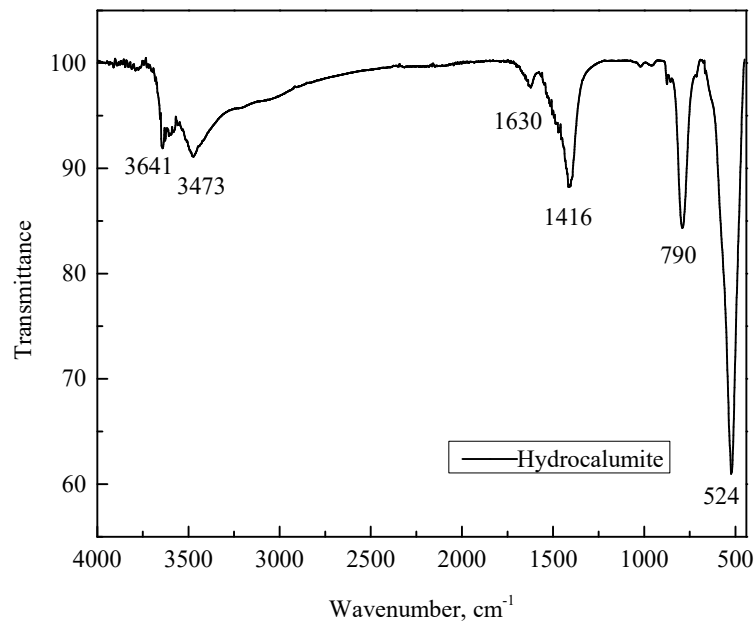


Figure 2. The FT-IR spectrum of the hydrocalumite synthesized in this work.

Figure 3 shows the SEM image of the synthesized hydrocalumite. The hydrocalumite samples show plate-like morphology and hexagonal crystallites with some obvious agglomeration.

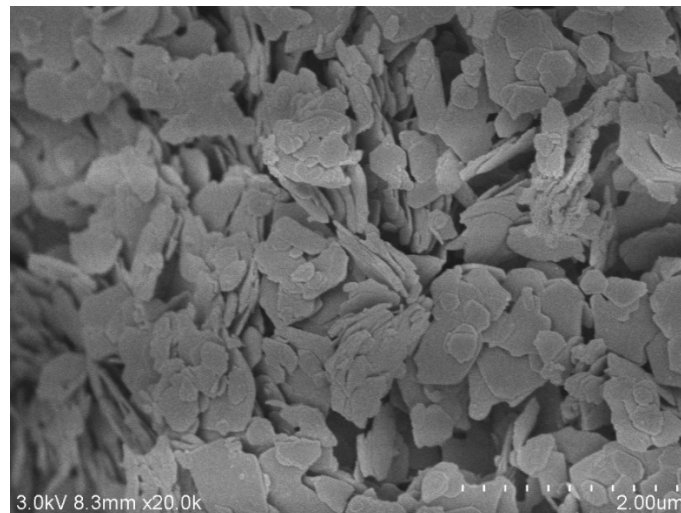


Figure 3. SEM morphologies of the hydrocalumite synthesized in this work.

3.2. Effect of Hydrocalumite Dosage on the Removal of the Silicate(IV) from Leaching Solution

The effect of hydrocalumite dosage on the removal of silicate(IV) was investigated by varying hydrocalumite dosage from 20 to 60 g/L. Figure 4 shows the influence of hydrocalumite dosage on the removal of silicate(IV) at 50 °C. In these experiments, the reaction time was fixed at 2 h. The removal percentage of silicate(IV) increased greatly to completeness as hydrocalumite dosage rose to 30 g/L and then began to decrease with the further increase of hydrocalumite dosage to 60 g/L. Unlike silicate(IV), no aluminate(III) was removed and thus it was possible to obtain pure aluminate(III) solution by selective adsorption of silicate(IV). The decrease in the removal percentage of silicate(IV) at a high dosage of hydrocalumite (40–60 g/L) might be ascribed to the agglomeration among the hydrocalumite, which reduces the surface area of adsorbents and blocks the adsorption sites [18,19]. Thus, an optimum

dosage of the hydrocalumite for the silicate(IV) removal was found to be 30 g/L. Therefore, 30 g/L of hydrocalumite was employed in the following experiments.

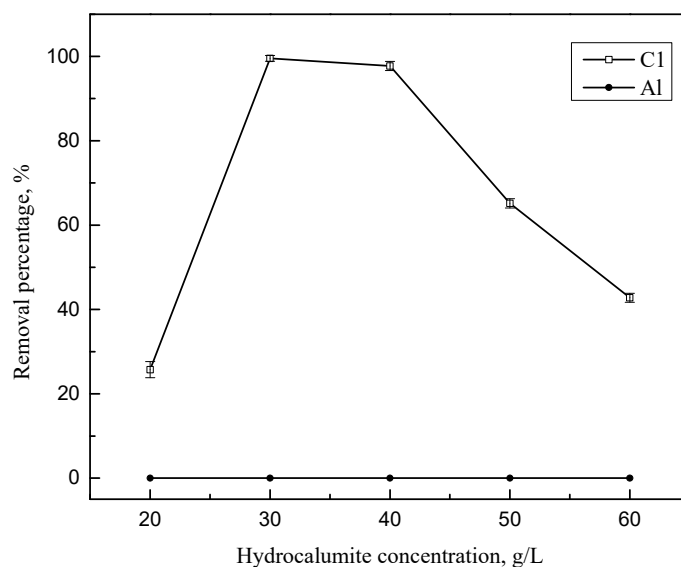
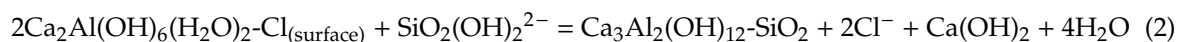


Figure 4. The effect of hydrocalumite dosage on the removal of the aluminate(III) and silicate(IV) from the solution. (Reaction time: 2 h, Temperature: 50 °C, Stirring speed: 400 rpm).

In 5 M NaOH solution, silicate(IV) exists as $\text{H}_2\text{SiO}_4^{2-}$ ($\text{SiO}_2(\text{OH})_2^{2-}$) and thus the adsorption reaction of silicate(IV) onto hydrocalumite can be represented as



First, silicates(IV) are adsorbed onto the surface by replacing the chloride ion on the surface of hydrocalumite. These adsorbed silicates(IV) $\text{H}_2\text{SiO}_4^{2-}$ are then intercalated into the interlayer via exchanging with the chloride ion in the interlayer as illustrated in Figure 5. The removal of silicate(IV) might occur at both the surface and interlayer sites through anion exchange. The intercalation of silicates(IV) leads to the phase transformation of hydrocalumite.

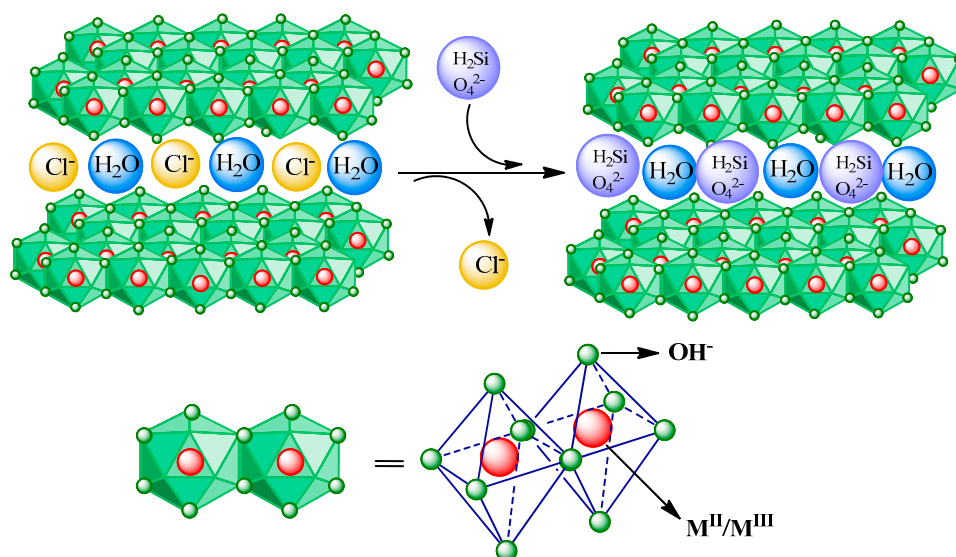


Figure 5. A schematic representation of silicate(IV) removal by adsorption onto hydrocalumite.

To elucidate the interaction mechanism between silicate(IV) and hydrocalumite, XRD characterizations of hydrocalumite after adsorption were conducted and the results are shown in Figure 6. Compared to Figure 1, the phase of hydrocalumite changed greatly after adsorption. Besides that, the morphology of hydrocalumite after the adsorption of silicate(IV) was studied by scanning electron microscopy (see Figure 7). It can be observed that the shape of hydrocalumite changed from plate-like morphology and hexagonal crystallites to polyhedral crystal block. XRD and SEM images indicated that after adsorption hydrocalumite exhibits the phase $\text{Ca}_3\text{Al}_2(\text{OH})_{12}\text{-SiO}_2$.

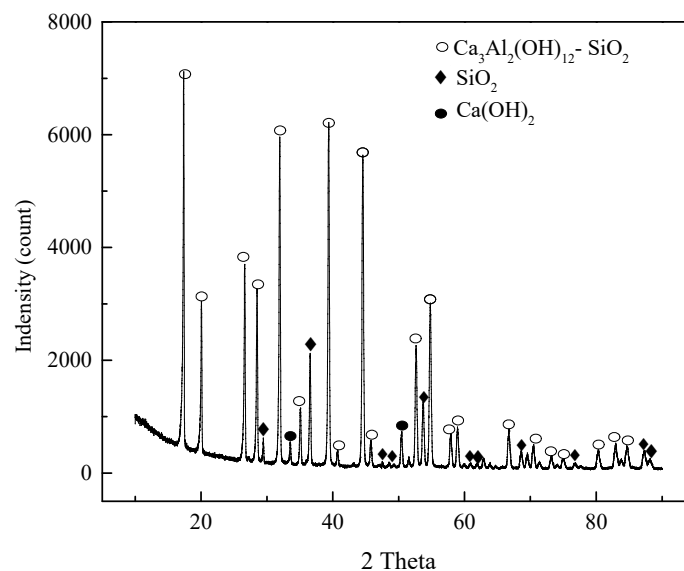


Figure 6. The XRD pattern of the hydrocalumite after removal of silicate(IV).

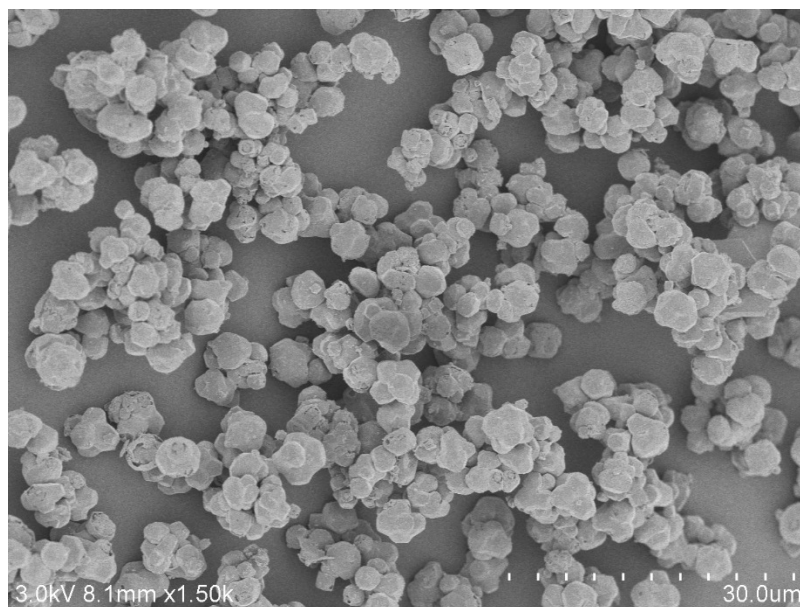


Figure 7. SEM images of hydrocalumite after adsorption.

3.3. Effect of Temperature

The effect of the reaction temperature on the removal of silicate(IV) was investigated at hydrocalumite dosage condition of 30 g/L. Figure 8 shows the effect of temperature on the removal of silicate(IV) in the temperature range from 20 to 90 °C. As the reaction temperature increased from 20 to 50 °C, the removal percentage of silicate(IV) rose greatly and most of the silicate(IV) was removed

at 50 °C. In the temperature range from 50 to 70 °C, silicate(IV) was completely removed from the solution. The decrease in the removal percentage of silicate(IV) beyond 70 °C may be attributed to the reaction enthalpy of the adsorption reaction. In general, the adsorption reaction is exothermic and thus desorption would occur at high temperature. Consequently, the adsorbed silicates(IV) might be desorbed when the reaction temperature rose to 90 °C. Another possibility is the instability of the hydrocalumite at 90 °C and thus the hydrocalumite might be inactivated [20]. This is in good agreement with the data that the hydrocalumite is stable below 85 °C [21,22].

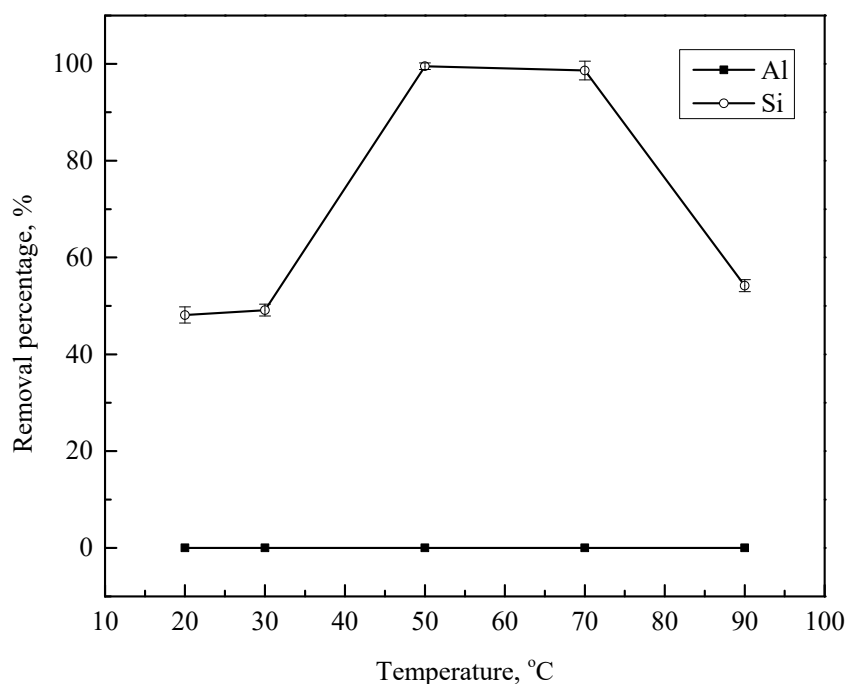


Figure 8. The effect of reaction temperature on the removal of the aluminate(III) and silicate(IV) from the solution. (Reaction time: 2 h, hydrocalumite dosage: 30 g/L, Stirring speed: 400 rpm).

3.4. Effect of the Reaction Time

The effect of the reaction time was investigated using 30 g/L hydrocalumite at 50 °C. As the reaction time increased from 30 mins to 4 h, the removal percentage of silicate(IV) slightly decreased from 100% to 90% (see Figure 9). When reaction time was less than 2 h, more than 20% of aluminate(III) was also removed with silicate(IV). However, the removal of aluminate(III) rapidly decreased when the reaction time was 2 h. Both $\text{H}_2\text{SiO}_4^{2-}$ and $\text{Al}(\text{OH})_4^-$ ions can be exchanged with the chloride ion in the interlayer of the hydrocalumite. When the reaction time was less than 2 h, aluminate(III) can be adsorbed onto hydrocalumite together with silicate(IV) owing to mass action effect. However, the adsorbed aluminate(III) ions could be exchanged with the silicate(IV) and thus the removal percentage of aluminate(III) decreased to negligible as reaction time reached to 2 h.

It has been reported that the adsorption of silicate(IV) onto hydrocalumite occurs more readily than that of aluminate(III) [23]. The adsorption of anions onto hydrocalumite occurs when the material is intercalated by weak electrostatic interactions with the layers. The exchange degree between silicate(IV) and aluminate(III) with the layers depends on the charge density of each anion. Therefore, the anion with higher charge density has a stronger electrostatic interaction with the layers [24]. In this case, the charge density of silicate(IV) ($970 \text{ C}\cdot\text{mm}^{-3}$) is much higher than that of aluminate(III) ($364 \text{ C}\cdot\text{mm}^{-3}$) [25]. At equilibrium, $\text{H}_2\text{SiO}_4^{2-}$ ions are strong enough to push $\text{Al}(\text{OH})_4^-$ ions out of the interlayer of hydrocalumite. At that time, all available sites might be covered and no active site is available for loading $\text{Al}(\text{OH})_4^-$ ions [26]. Table 1 shows the concentration of Al(III) and Si(IV) after the

specified adsorption treatments. The removal percentage of Al(III) and Si(IV) in Figures 4, 8 and 9 was calculated on the basis of this data.

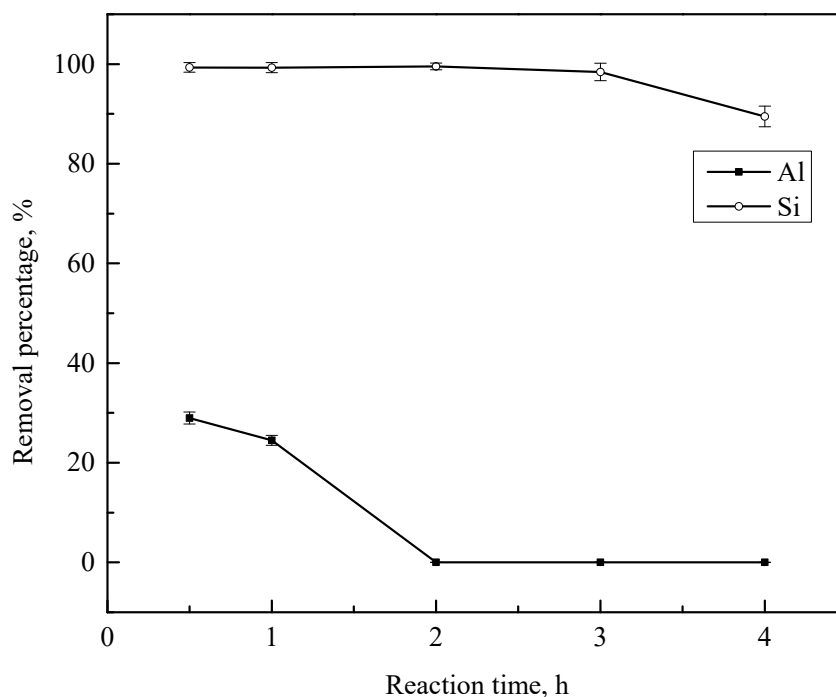


Figure 9. The effect of the reaction time on the removal of the aluminate(III) and silicate(IV) from the solution. (hydrocalumite dosage: 30 g/L, Stirring speed: 400 rpm, temperature: 50 °C).

Table 1. The concentration of Al(III) and Si(IV) in the solution after adsorption treatment with hydrocalumite. (The initial concentration of Al(III) and Si(IV) was 14,954.8 ppm and 150 ppm).

Conditions	Al(III), ppm	Si(IV), ppm
Effect of Hydrocalumite concentration		
20 g/L	15,094	111.4
30 g/L	15,028	0.6
40 g/L	15,291.4	3.4
50 g/L	15,590.8	52.2
60 g/L	15,690.9	85.9
Effect of temperature		
20°C	15,690.8	77.8
30°C	15,594.1	76.3
50°C	15,028	0.6
70°C	15,291.4	2.1
90°C	15,299.8	68.7
Effect of reaction time		
0.5 h	10,622.1	1.0
1 h	11,291.6	1.1
2 h	15,028.0	0.6
3 h	15,253.4	2.4
4 h	15,356.5	15.8

Our data indicated that the interaction between silicate(IV) and hydrocalumite depends on the hydrocalumite dosage, temperature and reaction time. At this condition, the purity of the aluminate(III) solution after the removal of silicate(IV) was higher than 99.99%. The change in the concentration of Al(III) and Si(IV) before and after adsorption together with the purity of Al(III) are shown in Table 2.

The data in Table 2 was obtained from the following conditions: 30 g/L hydrocalumite, 50 °C, 2 h, 400 rpm.

Table 2. Change in the concentration of Al(III) and Si(IV) in the solution and the corresponding purity of Al(III) before and after adsorption by hydrocalumite.

Composition	Al(III)	Si(IV)	Purity of Al(III) (%)
Before adsorption (ppm)	14,954.8	150	99
After adsorption (ppm)	15,028.0	0.6	99.99

3.5. Effect of Silicate(IV) Concentration

Figure 10 shows the effect of the initial concentration of silicate(IV) in the solution on their removal by adsorption onto hydrocalumite. In these experiments, the reaction time was fixed at 2 h. As the initial concentration of silicate(IV) increased from 150 to 900 mg/L, the removal percentage of silicate(IV) decreased a little. This decrease is related to the dosage of hydrocalumite. In these experiments, aluminate(III) was not adsorbed onto hydrocalumite and thus a pure aluminate(III) solution can be obtained.

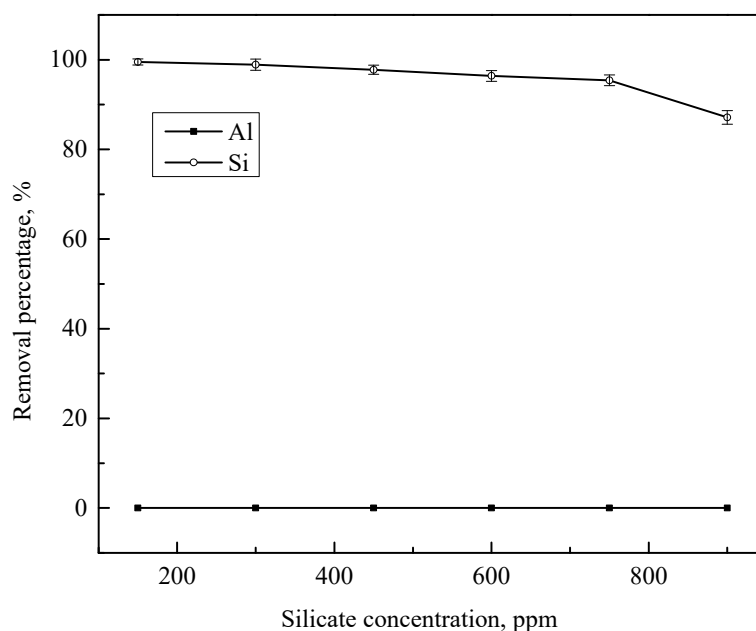


Figure 10. The effect of silicate(IV) concentration on the removal of the aluminate(III) and silicate(IV) from the solution. (hydrocalumite dosage: 30 g/L, Stirring speed: 400 rpm, temperature: 50 °C).

3.6. Adsorption Isotherm

The adsorption isotherm of silicate(IV) onto hydrocalumite was identified by applying Langmuir [27] and Freundlich [28] isotherms to our adsorption data.

$$Q_e = \frac{Q_m K_L C_e}{1 + b C_e} \quad (\text{Langmuir isotherm}) \quad (3)$$

$$\ln Q_e = \ln k_f + \frac{1}{n} \ln C_e \quad (\text{Freundlich isotherm}) \quad (4)$$

where Q_e is the amount of metal ions adsorbed at equilibrium (mg/g), C_e is the equilibrium concentration (mg/L), Q_m is the maximum adsorption capacity (mg/g), K_L is the Langmuir constant (L/mg), k_f and $1/n$ are the Freundlich constants. Figure 11 shows the results on the application of both isotherms to the adsorption data. The correlation coefficients (R^2) of the Freundlich isotherm was higher than that

of Langmuir, indicating that the adsorption of silicate(IV) onto hydrocalumite follows the Freundlich isotherm.

Table 3 lists the physical constants for the Freundlich isotherm obtained from Figure 11. The Freundlich isotherm can explain the adsorption occurring on the heterogeneous surfaces or surface supporting sites of varied affinities. The value of $1/n_F$ (0.29) indicates that the adsorption of silicate(IV) onto hydrocalumite occurs readily [29]. The adsorption onto the surface of hydrocalumite can occur easily but the exchange within the interlayer anions needs diffusion. Therefore, the adsorption onto hydrocalumite is a complex heterogeneous process and thus the Freundlich isotherm is better at explaining the adsorption of silicate(IV) onto hydrocalumite.

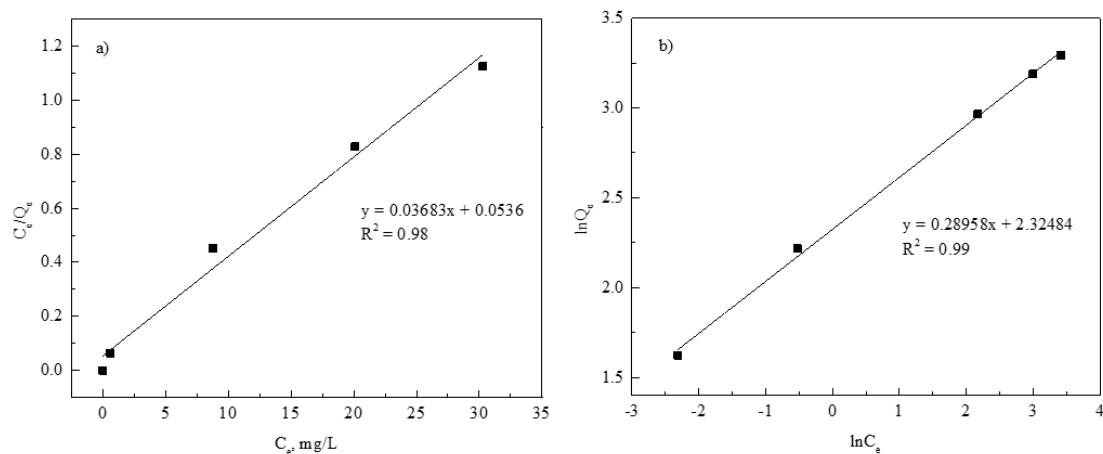


Figure 11. A comparison of the adsorption isotherm of silicate onto hydrocalumite between the Langmuir (a) and Freundlich isotherms (b).

Table 3. Estimated parameters for Freundlich adsorption isotherm from the adsorption data.

Ion	Freundlich		
	$1/n_F$	K_F	R^2
Si	0.29	10.23	0.99

4. Conclusions

The NaOH leaching solution of mechanically activated aluminum dross contains a small amount of silicate(IV) with a major component of aluminate(III). In order to recover pure aluminate(III) solution, the removal of silicate(IV) by reaction with hydrocalumite was investigated. Successful synthesis of hydrocalumite was verified via XRD, SEM images and FTIR. In this work, the effect of hydrocalumite dosage, temperature, reaction time and silicate(IV) concentration were studied. When reaction time was less than 2 h, about 20% of the aluminate(III) was loaded onto hydrocalumite. Therefore, at least 2 h of reaction time is necessary to reduce the loss of aluminate(III) owing to co-adsorption. When the dosage of hydrocalumite was high compared to the concentration of silicate(IV), the removal percentage of silicate(IV) was rather reduced. The reaction temperature of 50–70 °C was adequate to the complete removal of silicate(IV). At the optimum conditions (30 g/L hydrocalumite, 50 °C, 2 h, 400 rpm), most of the silicate(IV) had been removed from the leaching solution. The adsorption of silicate(IV) followed the Freundlich isotherm.

Author Contributions: M.S.L. designed the research and helped analyze data. T.T.N.N. performed experiments and wrote the paper.

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Conflicts of Interest: The authors declare no conflict of interest.

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