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Adsorption of Cadmium(II) Using Ca/Al Layered Double Hydroxides Intercalated with Keggin Ion

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Abstract: Ca/Al layered double hydroxides (Ca/Al LDH) was synthesized using co-precipitation method following calcination at 800 °C and was intercalated with Keggin ion $[\alpha\text{-SiW}_{12}\text{O}_{40}]^{4-}$ to form intercalated Ca/Al LDH. Materials were characterized using XRD and FTIR spectrophotometer. Furthermore, materials were used as an adsorbent of cadmium(II) from solution. The results showed that layer material was formed completely after calcination which was indicated at diffraction 20° due to loss of water in the interlayer space. Ca/Al LDH after calcination was intercalated with $[\alpha\text{-SiW}_{12}\text{O}_{40}]^{4-}$ ion and interlayer distance was increased from 4.25 to 4.41 Å showed that intercalation process was successfully conducted. Adsorption of cadmium(II) using Ca/Al LDH was conducted at pH 9 and intercalated Ca/Al LDH at pH 8 showed that intercalated material has slightly faster than Ca/Al LDH without intercalation probably due to slightly increasing interlayer distance of Ca/Al LDH after intercalation. The adsorption capacity of intercalated Ca/Al LDH was higher than Ca/Al LDH without intercalation at the temperature range of 30–50 °C.

Keywords: Ca/Al LDH; intercalation; $[\alpha\text{-SiW}_{12}\text{O}_{40}]^{4-}$; adsorption; cadmium(II)

■ INTRODUCTION

Basically, the presence of various heavy metal in the environment is essential for supporting the life of any living creatures. However, the entering of many heavy metals into the open environment due to the effluent discharge from various anthropogenic activities leads to numerous environmental problems [1]. Cadmium is one of the most toxic heavy metals caused by its high tendency for bioaccumulation and high mobility in the environment [2]. Moreover, cadmium is classified as the top ten of the most dangerous chemicals for human health and it cannot naturally be removed or reduces. According to the World Health Organization (WHO), the limit of permissible cadmium concentration in the groundwater

is less than 3 µg/L. Even though, cadmium is still widely used in many industrial process such as batteries, color pigment, and electroplating [2]. Hence the removal of cadmium from aqueous solution is a mandatory in order to maintain the environmental live cycles and to support the green environment process.

Until the recent decade, numerous attempts in the heavy metal reduction and removal from the environment have been carried out all over the world. Various physical and chemical methods have been developed like oxidation of metal, filtration, coagulation, membrane separation, and also adsorption [3-4]. Among these methods, adsorption is considered as the most suitable to remove contaminant of heavy metals from wastewater [5]. The successful adsorption process

depends on the properties and capability of the used adsorbent. Various adsorbents have been used for adsorption of heavy metals such as zeolites, clay materials, bentonite, kaolin, metal oxides, layered double hydroxides, and also organic adsorbent [6-9].

Layered double hydroxides (LDH) is inorganic layer materials consisting of divalent (M^{2+}) and trivalent (M^{3+}) metal cations with anion in interlayer space. The general formula of LDH is given as $[M^{2+}_x M^{3+}_y (OH)_2]^{3+} (A^{n-})_{x/n} \cdot nH_2O$, where M is divalent and trivalent metal ions and A^{n-} is interlayer anions with valence n [10-11]. Common divalent cations for the synthesis of LDH are group 2, Cu^{2+} , Zn^{2+} , and divalent transition metals. Trivalent cations are Al^{3+} , Mn^{3+} , Fe^{3+} , Co^{3+} , and Cr^{3+} and also other trivalent transition metals. Anion in the interlayer space is commonly from a synthetic solution such as nitrate, chloride, and carbonate [12]. The unique properties of LDH are flexibility, where anion in the interlayer space can be exchangeable depending on the application of LDH. Exchangeable anion in the interlayer distance is like a guest of materials and easily to exchange in order to obtain unique properties of LDH. LDH has been applied for adsorbent of heavy metal ions such as Pb^{2+} [13], Cu^{2+} [14], In^{3+} [15], and Cd^{2+} [10] from aqueous solution.

Polyoxometalates are early transition metal clusters consists of addenda (W, Mo, V, etc) and heteroatoms (P, Si, B, Co, etc) [16-17]. Polyoxometalates have various size, shape, and also have a unique solubility, acid-base, and redox properties. The solubility of polyoxometalates is depending on counterions to form large anion clusters [18-19]. Recently, polyoxometalate anion has been widely used as large anion to be inserted into the interlayer space of LDH to form materials with unique properties for various applications such as adsorption and catalysis. Polyoxometalates with Keggin, Dawson, and Anderson types are frequently used as anion for intercalating the interlayer LDH to increase its interlayer distance.

In order to enhance the adsorption capacity of Ca/Al LDH, here we have applied the Keggin-type of $[\alpha-SiW_{12}O_{40}]^{4-}$ polyoxometalate to be intercalated into Ca/Al LDH. As reported by Ma et al., intercalation of $[PW_{12}O_{40}]^{3-}$ polyoxometalate onto Mg-Al LDH has

successfully increased its surface area [20]. Herein, we report the synthesis of Ca/Al LDH intercalated by $[\alpha-SiW_{12}O_{40}]^{4-}$ polyoxometalate which has a similar structure with $[PW_{12}O_{40}]^{3-}$ namely Keggin ion using coprecipitation followed by ion exchange method. The product was then utilized as an adsorbent in the removal of toxic heavy metal i.e., Cd^{2+} , from aqueous solution. For the comparison, Ca/Al LDH without intercalated with polyoxometalate was also prepared as tested for Cd^{2+} adsorption.

EXPERIMENTAL SECTION

Materials

Chemicals such as sodium tungstate ($Na_2WO_4 \cdot 2H_2O$), hydrochloric acid (HCl), sodium metasilicate (Na_2O_3Si), potassium chloride (KCl), sodium carbonate (Na_2CO_3), sodium hydroxide (NaOH), potassium hydroxide (KOH), sodium chloride (NaCl), cadmium(II) nitrate ($Cd(NO_3)_2$), calcium nitrate ($Ca(NO_3)_2 \cdot 4H_2O$), and aluminum(III) nitrate ($Al(NO_3)_3 \cdot 9H_2O$) were obtained from Merck and Sigma Aldrich and directly used without further purification with grade > 99%. Water was purified using cycling ion exchange process by Purite® water system technology at pH 7 at an integrated research laboratory, graduate school, Universitas Sriwijaya.

Instrumentation

The analysis was conducted using a Shimadzu FTIR Prestige-21 spectrophotometer. The sample was mixed with KBr to form KBr pellet. The measurement was performed at wavenumber $400-4000\text{ cm}^{-1}$. Analysis of XRD was conducted using X-Ray powder diffraction Rigaku Miniflex-600. The sample was scanned at speed scan $0.1^\circ\text{ min}^{-1}$. Adsorption of cadmium(II) was determined using a UV spectrophotometer EMC-61PC double beam spectrophotometer by complexation using 1,10-phenanthroline.

Procedure

Synthesis of Ca/Al LDH

Synthesis Ca/Al layered double hydroxides was conducted according to Granados-Reyes et al. (2017) by the co-precipitation method with slight modification

[21]. Calcium nitrate and aluminum nitrate with molar ratio 2:1 ($\text{Ca}^{2+}/\text{Al}^{3+}$) were prepared by adding sodium hydroxide 2 M. The mixtures were stirred at 60 °C and pH was adjusted to 11 by adding sodium hydroxide. The mixtures were stirring until pH constant. After stable pH, the mixtures were heated using autoclave at 120 °C for 2 h. The mixtures were filtered at room temperature and washed with water and dried at 80 °C overnight to form white solid Ca/Al layered double hydroxides. Calcination of Ca/Al layered double hydroxides was performed at 800 °C for 3 h with a heating rate of 300 °C/h. Characterization was performed using XRD and FTIR analyses.

Synthesis of Keggin ion [$\alpha\text{-SiW}_{12}\text{O}_{40}$]⁴⁻ [22]

Keggin ion of [$\alpha\text{-SiW}_{12}\text{O}_{40}$]⁴⁻ was obtained as potassium salt $\text{K}_4[\alpha\text{-SiW}_{12}\text{O}_{40}]$ as follows using sodium salts of starting materials. Sodium metasilicate (11.0 g) was dissolved into 100 mL of water (solution A). Sodium tungstate (182.0 g) was dissolved into 300 mL of boiling water (solution B). A solution of 4 M HCl (165 mL) was added drop by drop to solution A over 5 min with vigorous stirring in order to dissolve the local precipitate of tungstic acid. Then solution A was added quickly to solution B followed by adding 50 mL of 4 M HCl. The pH was adjusted to 6. The solution was kept at 373 K for 1 h. A solution of 1 M sodium tungstate (50 mL) and immediately thereafter, 80 mL of 4 M HCl were added. The solution was filtered after cooling to room temperature. The potassium salt of $\text{K}_4[\alpha\text{-SiW}_{12}\text{O}_{40}]$ was obtained by adjusting pH to around 2 with aqueous 1 M KOH and then solid KCl (50 g) was added. A white precipitate of $\text{K}_4[\alpha\text{-SiW}_{12}\text{O}_{40}]$ was obtained. $\text{K}_4[\alpha\text{-SiW}_{12}\text{O}_{40}]$ was characterized using FT-IR spectrophotometer.

Intercalation Ca/Al LDH with Keggin ion of [$\alpha\text{-SiW}_{12}\text{O}_{40}$]⁴⁻

Intercalation of Ca/Al layered double hydroxides with [$\alpha\text{-SiW}_{12}\text{O}_{40}$]⁴⁻ was carried out according to work done by Ma et al. [20] with slight modification using an ion exchange method as follows. Polyoxometalate of $\text{K}_4[\alpha\text{-SiW}_{12}\text{O}_{40}]\cdot n\text{H}_2\text{O}$ (1.0 g) was dissolved with 50 mL of water. Ca/Al layered double hydroxides (2 g) was mixed

with 25 mL of 1 M sodium hydroxide. Into this mixture was added a solution of polyoxometalate with gentle stirring under nitrogen condition. The reaction was performed at 24 h to form a white suspension. The white suspension was filtered by dry vacuum and wash with water several times. The solid material was kept at 110 °C for 2 days. The product of intercalated Ca/Al with [$\alpha\text{-SiW}_{12}\text{O}_{40}$]⁴⁻ was analyzed using XRD powder analysis and identification using FTIR spectroscopy.

pH PZC analysis

The pH Point Zero Charges (pH PZC) were measured using a solution of sodium chloride 0.1 M as follows. Sodium chloride 0.1 M was adjusted from pH 1 to 10 by addition of sodium hydroxide or hydrochloric acid 0.1 M. In the series of sodium chloride solution with various pH, about 1.0 g of Ca/Al layered double hydroxides was added. The mixtures were kept for 24 h in constant stirring. Mixtures were filtered and pH of the filtrate was determined by pH meter. pH PZC graph was obtained by comparison initial and final pH solution.

Adsorption studies

Adsorption of cadmium(II) using Ca/Al LDH and intercalated Ca/Al was performed by the studied effect of pH, adsorption time, the initial concentration of cadmium(II), and temperature. The acidity of adsorption was investigated by varying at pH 1, 2, 3, 4, 5, 6, 7, and 8. Adsorption time was studied by varying the time at 10, 20, 30, 40, 50, 60, and 90 min using 0.1 g of adsorbent. Effect initial concentration of cadmium(II) and the temperature was studied using concentration of 10, 20, 30, 40, 50, and 60 mg/L at 30, 40, and 50 °C. The filtrate of the solution after adsorption was complexed with 1,10-phenanthroline and was analyzed using UV at λ 326 nm. The adsorption performance in term of adsorption capacity (q_e) and adsorption efficiency can be calculated by the following equation.

$$q_e = \frac{(C_o - C_t)}{m} \cdot V \quad (1)$$

$$\% \text{ Removal} = \frac{(C_o - C_t)}{C_o} \times 100 \quad (2)$$

■ RESULTS AND DISCUSSION

Characterization of Ca/Al LDH was performed using XRD powder analysis as shown in Fig. 1(a). Specific diffraction of Ca/Al LDH was reported by Granados-Reyes et al. [21] in which diffraction appeared at 10–50°. Synthesized Ca/Al LDH as shown in Fig. 1(a) has small diffraction at 10° and high diffraction at 20–30° indicated the crystalline phase of Ca/Al LDH due to nitrate and carbonate. The presence of nitrate and carbonate can be explained by synthetic materials because of atmospheric air conditions. This anion was located between interlayer of Ca/Al LDH. Calcination was performed at 800 °C in the air at 3 h and diffraction was shown in Fig. 1. The intensity of calcite, which was recorded at 2θ around 30° [23], increased by increasing temperature. Other phases were detected as Katoite at 17.3° [211], 20.11° [220], 28.5° [321], 31.9° [400], 36.58° [420], 39.3° [431], 44.6° [521], 52.8° [611], 55.0° [640], 57.74° [642], mayenite at 53° [640], and 56° [642] (JCPDS24-0217) [24]. The concentration of calcite was decreased to 800 °C because this compound decomposes at a lower temperature than mayenite and calcium oxide. Layer materials of Ca/Al was identified as mayenite $\text{Ca}_{12}\text{Al}_4\text{O}_{33}$. The interlayer distance of Ca/Al LDH was identified at diffraction 20° [220] resulted in gallery 4.25 Å.

Polyoxometalate $\text{K}_4[\alpha\text{-SiW}_{12}\text{O}_{40}]\cdot n\text{H}_2\text{O}$ is a crystalline material. As shown in Fig. 1, the X-ray diffraction pattern of the synthesized Keggin polyoxometalate exhibited

several sharp diffraction peaks at $2\theta = 7\text{--}10^\circ$, $16\text{--}22^\circ$, $25\text{--}30^\circ$, and $32\text{--}38^\circ$. This finding revealed that the synthesized material is appropriate with the Keggin structure since these diffraction peaks are the typical diffraction pattern of Keggin polyoxometalate [25]. Intercalation of Ca/Al LDH with $\text{K}_4[\alpha\text{-SiW}_{12}\text{O}_{40}]\cdot n\text{H}_2\text{O}$ resulted in diffraction as indicated in Fig. 1. Keggin ion of $[\alpha\text{-SiW}_{12}\text{O}_{40}]^{4-}$ was identified at diffraction 9° with small intensities (24.71 cts). Layered material of Ca/Al was detected at diffraction 20° with high intensities. Several diffractions were detected at 30, 32, 35, 36, and 40° due to insertion of Keggin ion to replace nitrate and water molecules.

After intercalation process, the Miller indices [220] peak was shifted to the lower 2θ value, particularly from 20.88° to 20.11° producing the increase of the $d(220)$ basal spacing from 4.25 Å to 4.41 Å. Although there was a slight increase of the interlayer distance, this finding is an indication that the intercalation process was successfully conducted. The diameter of Keggin ion is 9.8 Å [20,26], thus intercalation process will increase gallery around ± 0.16 Å. Intercalation process in this experiment showed that the position of Keggin ion probably in a diagonal position, not in a vertical position as illustrated in Fig. 2.

Identification using FTIR spectroscopy was shown in Fig. 3. Unique vibrations of Ca/Al LDH as identified in Fig. 3(a) were identified at wavenumber 660 and 790 cm^{-1} ,

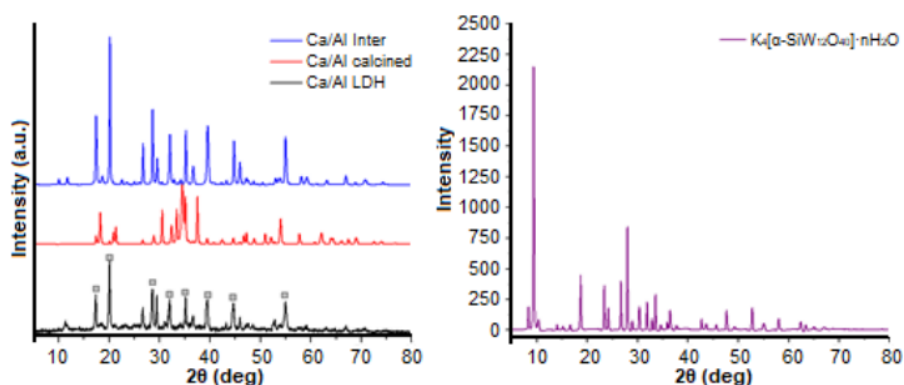


Fig 1. XRD powder patterns of Ca/Al LDH, calcination of Ca/Al LDH at 800 °C, polyoxometalate $\text{K}_4[\alpha\text{-SiW}_{12}\text{O}_{40}]\cdot n\text{H}_2\text{O}$ and intercalated Ca/Al LDH with $\text{K}_4[\alpha\text{-SiW}_{12}\text{O}_{40}]\cdot n\text{H}_2\text{O}$



Fig 2. The plausible position of $[\alpha\text{-SiW}_{12}\text{O}_{40}]^{4-}$ toward Ca/Al LDH

which were assigned as Ca-O and Al-O vibrations, respectively. Generally, calcination by increasing temperature creates sharp vibration at $700\text{--}800\text{ cm}^{-1}$. We observed small vibration after calcination due to water and nitrate ions between interlayer space is still remains probably due to hydrogen bonding. The vibration of OH at 3300 cm^{-1} was also to be sharpened due to loss of water and OH vibration is assigned as bonding OH with metal M^{2+}/M^{3+} [27]. On the other hand, the vibration of nitrate at 1380 cm^{-1} was a broader and split with increasing calcination temperature due to the stability of nitrate ion toward high temperature (Fig. 3(b)). Polyoxometalate $K_4[\alpha\text{-SiW}_{12}\text{O}_{40}]\cdot n\text{H}_2\text{O}$ as indicated in Fig. 3(c) has the specific vibration of Si-O at 926 cm^{-1} , W=O at 980 cm^{-1} , W-Oc-W at 881 cm^{-1} , and W-Oe-W at 780 cm^{-1} . Intercalation of Ca/Al LDH with Keggin ion of $[\alpha\text{-SiW}_{12}\text{O}_{40}]^{4-}$ has a broad vibration at 1472 cm^{-1} . These vibrations were shifted to higher wavenumber (1400 to 1472 cm^{-1}) due to Keggin ion replace nitrate ion in Ca/Al LDH [28]. Thus, intercalation of Ca/Al LDH using intercalant $[\alpha\text{-SiW}_{12}\text{O}_{40}]^{4-}$ was successfully conducted. For further utilization, the material was used as an adsorbent of cadmium (II) from aqueous solution.

Stability of Ca/Al LDH and intercalated Ca/Al LDH was firstly identified before adsorption process using pH PZC. The results of pH PZC for Ca/Al LDH and intercalated Ca/Al LDH are given in Fig. 4.

The graph of pH PZC in Fig. 4 showed that the intersection point was appeared at pH 9 for Ca/Al LDH and at pH 8 for intercalated Ca/Al LDH. Both Ca/Al LDH and intercalated Ca/Al LDH are base materials. pH PZC indicated zero charges of material. There are no both positive and negative charges at that points [29]. Based on adsorption data, adsorption of cadmium(II) was found optimum at pH 4 for Ca/Al LDH and at pH 3 for intercalated Ca/Al LDH. These results were different from

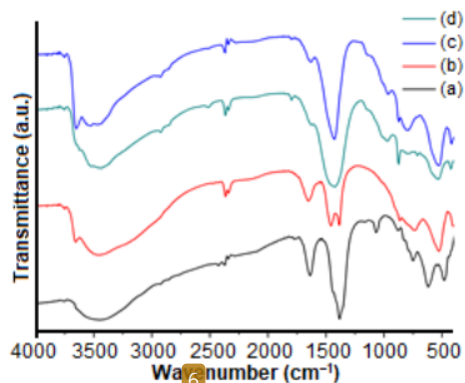


Fig 3. FTIR spectrum of (a) Ca/Al LDH, (b) calcination of Ca/Al LDH at $800\text{ }^\circ\text{C}$, (c) polyoxometalate $K_4[\alpha\text{-SiW}_{12}\text{O}_{40}]\cdot n\text{H}_2\text{O}$, and (d) intercalated Ca/Al LDH with $K_4[\alpha\text{-SiW}_{12}\text{O}_{40}]\cdot n\text{H}_2\text{O}$

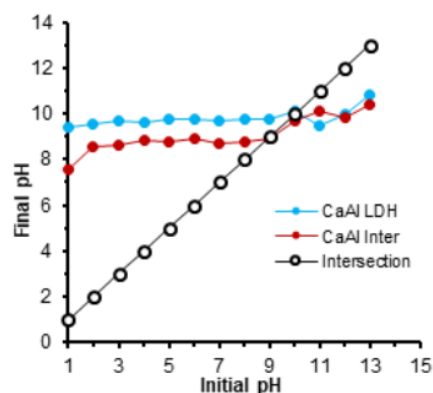


Fig 4. pH PZC graph

pH PZC analysis due to acid properties of Cd(II) on a solution as reported by Jancsó [30]. Cadmium existed in the solution below pH 9 and all cadmium is precipitate as hydroxide at pH above 9. Thus, the adsorption experiment was conducted at pH 4 for adsorbent Ca/Al LDH and pH 3 for adsorbent intercalated Ca/Al LDH.

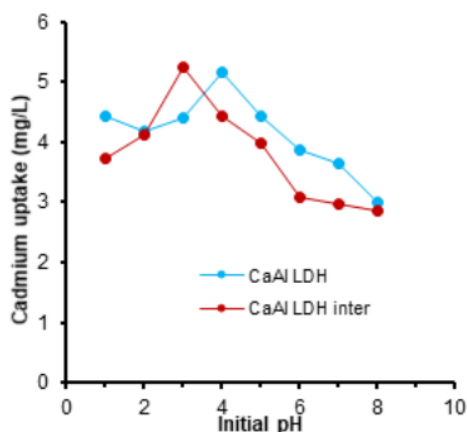


Fig 5. Effect of pH on adsorption of Cd(II) onto Ca/Al LDH and intercalated Ca/Al LDH

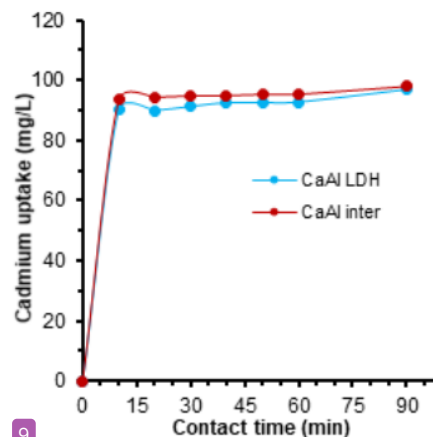


Fig 6. Effect of adsorption time of Cd(II) on Ca/Al LDH and intercalated Ca/Al LDH

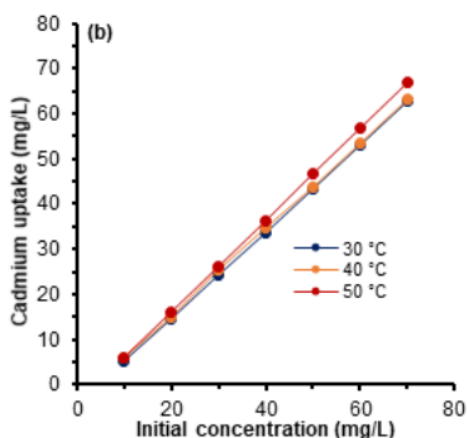
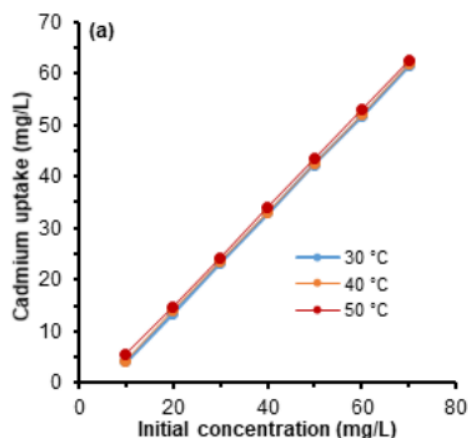


Fig 7. Effect of initial concentration of cadmium (II) and temperature on adsorption using (a) Ca/Al LDH and (b) intercalated Ca/Al LDH

Adsorption of cadmium(II) was first investigated by studying the effect of adsorption time as shown in Fig. 6. The adsorption profile of cadmium(II) showed that adsorption was faster at an initial time for intercalated Ca/Al LDH than Ca/Al LDH. The amount of adsorption was increased sharply by increasing adsorption time. The adsorption rate of data in Fig. 6 was calculated using the Langmuir-Hinshelwood equation [31]. The results showed that intercalated Ca/Al LDH and Ca/Al LDH have adsorption rate of 0.038 and 0.034 min^{-1} , respectively. These data showed that intercalated Ca/Al

LDH has reactivity slightly higher than Ca/Al LDH due to slightly increase interlayer distance after the intercalation process.

The further experiment of adsorption was conducted with a different initial concentration of cadmium(II) and temperature as shown in Fig. 7. Only slightly different effect of temperature adsorption in various concentration of cadmium(II). Data in Fig. 7 be calculated using the Langmuir equation [32], and the results are presented in Table I.

Data in Table I showed that increasing temperature

Table 1. Adsorption capacity of Ca/Al LDH and intercalated Ca/Al LDH

Layered Double Hydroxides	Temperature (°C)	Adsorption Capacity (mg/g ⁻¹)	R-Value
Ca/Al	30	0.260	0.9300
	40	0.945	0.9352
	50	1.125	0.9902
Intercalated Ca/Al	30	0.479	0.9406
	40	1.240	0.9198
	50	12.578	0.9272

would largely increase the adsorption capacity of LDH. The data also indicates intercalated Ca/Al LDH has higher adsorption capacity than Ca/Al without intercalation Keggin ion. Adsorption of cadmium(II) is largely increased at 50 °C on intercalated Ca/Al LDH probably because adsorption occurred not only on the surface of intercalated Ca/Al LDH but also onto interlayer distance. Thus, intercalation of Ca/Al LDH will give a significant effect for the adsorption of cadmium(II) in aqueous solution.

CONCLUSION

In this work, Ca/Al LDH intercalated with Keggin ion $[\alpha\text{-SiW}_{12}\text{O}_{40}]^{4-}$ was successfully prepared; it was proofed by increasing the interlayer distance of the Ca/Al LDH structure. Since the increase of the interlayer distance occurred in slight, we assumed that the insertion of the Keggin ion into the interlayer space was in diagonal position. The both Ca/Al LDH and intercalated Ca/Al LDH were tested to adsorb cadmium(II) ion from aqueous solution. According to the adsorption kinetic study, the adsorption of cadmium(II) onto intercalated Ca/Al LDH was faster rather than that on without intercalation one. Moreover, the adsorption capacity of intercalated Ca/Al LDH is higher indicating that the intercalated Ca/Al LDH is more effective adsorbent to adsorb cadmium(II) ion from aqueous solution.

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