



Removal of Arsenic from Synthetic Acid Mine Drainage using Mn-Fe Layered Double Hydroxide Adsorbent

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Abstract

The Mn-Fe layered double hydroxide using chloride in the interlayer anion was successfully synthesized using chemical co-precipitation methods. The Mn-Fe LDH was then applied as adsorbent for arsenic removal from synthetic acid mine drainage. The adsorbent characterizations of SEM and XRD analysis showed that the Mn-Fe LDH had many different functional groups and a high specific surface area for the adsorption processes. The morphological structure of Mn-Fe LDH by the SEM-EDS analysis method shows a round shape structure with a particle size of about 1 μm , and the XRF analysis method shows that the Mn and Fe elements dominate more than other components. Batch adsorption experimental conducted using the Mn-Fe LDH with the interlayer anion of chloride as an adsorbent to study the effect of contact time, equilibrium pH, and temperature on the arsenic removal. The Mn-Fe LDH showed high adsorption uptake capacity and selectivity for the arsenic in the synthetic acid mine drainage. The adsorption and ion exchange between interlayer chloride anions in Mn-Fe LDH and As (V) solution was the main adsorption mechanism. Therefore, the Mn-Fe LDH can be used as an adsorbent in water and wastewater treatment. In contrast, this research has the potential to be processed and developed into advanced materials.

Keywords: adsorption, acid mine drainage, co-precipitation, Mn-Fe LDH, wastewater treatment

1. Introduction

Acid mine drainage (AMD) is water generated from mining activities with a deficient pH level due to oxidation of sulfide minerals such as pyrite, which have resulted in a high concentration of dissolved sulfate and heavy metal contamination of surface and groundwater (Acharya and Kharel, 2020, Otgonjargal *et al.*, 2012, Ryu *et al.*, 2020). The AMD has a real impact on mining activities, including runoff and infiltration from mine rock, and it is still a problem that arises in the community due to the high contamination of dissolved sulphate content and heavy metals contamination such as As, Cd, Co, Cu, Ni, Pb, Zn and other metals dissolved in it (Kefeni *et al.*, 2017, Ryu *et al.*, 2020). The AMD has a negative impact on water quality both during mining and post-mining activities. South Kalimantan has many coal and mineral mining activities, which have resulted in the AMD impacts, including polluting community wells living around the mining area. The community's well water is used to meet the needs both for bathing and for consumption as drinking water. The continuous use and consumption of polluted

water due to AMD's impact causes these metals to be directly absorbed on the skin and accumulate in the body so that it can cause various health problems such as skin diseases, indigestion, and even the worst-case cause paralysis. In the AMD containing-As, As found as the inorganic oxyanions forms As (III) and As (V), whereas: As (III) is more harmful than As (V), and its removal is more complicated (Liu *et al.*, 2019, Otgonjargal *et al.*, 2012, Suvokhiaw *et al.*, 2016).

The World Health Organization (WHO) devalued the standard for arsenic in drinking water to 0.01 mg/L. As (V) can be easily eliminated by adsorption technology using adsorbents, but it is difficult to remove As (III) by adsorption processes, and therefore, As (III) requires oxidation before adsorption (Liu *et al.*, 2019, Mohan and Pittman, 2007, Otgonjargal *et al.*, 2012, Suvokhiaw *et al.*, 2016). There are many methods of removing As (V); among them, the adsorption method is regarded as high efficiency, relatively cost-effective technology, and minimum generation of toxic sludge for water treatment (Barrera *et al.*, 2017, Liu *et al.*, 2019, Núñez-Gómez *et al.*, 2019, Otgonjargal *et al.*, 2012,

Suvokhiaw *et al.*, 2016, Zhou *et al.*, 2018). Various adsorbents have been developed in previous studies, such as clay minerals, activated alumina, activated carbon, and ion exchange resin. Compared with the typical cation clay minerals, anionic clays are mostly in the form of layered double hydroxides (LDHs), including hydrotalcite and hydrotalcite-like (Liu *et al.*, 2019, Otgonjargal *et al.*, 2012, Xie *et al.*, 2019, Zubair *et al.*, 2017).

Layered double hydroxides (LDHs) are known as hydrotalcite-like compounds (HTLcs) are categorized of anionic clays consisting of stacked positively charged layers of metal hydroxides, separated and neutralized by exchangeable anions in the interlamellar galleries and some water molecules. The formula of LDH can be written as $[M^{2+}_{1-x}M^{3+}_x(OH)_2]^+(A^{n-}_{x/n}) \cdot mH_2O$, where M^{2+} and M^{3+} represent the di- (Mg, Mn, Ni, Co or Zn) and trivalent (Al, Fe, or Cr) metal ions and A^{n-} is the interlayer anion, such as carbonate and chloride or many various anions depending on the different synthesis routes (Balsamo *et al.*, 2012, Iwai *et al.*, 2019). LDHs are assembled by a non-covalent bond, where the positively charged main lamellar includes divalent or trivalent cations and non-framework interlayer anions along with water molecules. LDHs exist as natural minerals; however, they can quickly and economically be synthesized. LDHs have relatively interlayer spaces, high surface area, non-toxicity, high chemical, and thermal stability. Therefore, they exhibit high capability and compatibility to capture organic and inorganic anions and "memory effect" allows the reconstruction of the original hydrotalcite structure (Kentjono *et al.*, 2010, Vithanage *et al.*, 2020). LDHs have been studied on their application in vast areas, i.e. supercapacitors, catalysis, photochemistry, electrochemistry, polymerization, magnetization, biomedical science, and environmental application. Recently, many researchers have evaluated the ability of LDHs in the removal of inorganic contaminants, such as oxyanions; e.g., dichromate, phosphate, selenite, borate, arsenite, arsenate, selenate, and nitrate; and monoatomic anions; e.g., fluoride, chloride, bromide, and iodide from aqueous solutions through adsorption and ion exchange.

Moreover, LDHs are alkaline and positively charged due to the nature of metal cations and metal-O bond, which provide electrostatic interactions with anionic pollutants (Kentjono *et al.*, 2010, Zubair *et al.*, 2017). Even some researcher had been studied LDH as adsorbent, however, the

interlayer anion on the LDH using chloride still need to be explored to distinguished compared to carbonate anion, then the research on capacity of the Mn-Fe LDH with this interlayer anion for arsenic removal is limited. This research aims to study the effect of contact time, equilibrium pH, and temperature on the arsenic adsorption process of the Mn-Fe LDH with the interlayer anion of chloride as an adsorbent.

2. Methodology

2.1. Materials

The chemicals used in this experiment are manganese (II) chloride tetrahydrate ($MnCl_2 \cdot 4H_2O$) (Merck KGaA, Darmstadt, Germany), ferric chloride hexahydrate ($FeCl_3 \cdot 6H_2O$) (Merck KGaA, Darmstadt, Germany), hydrogen chloride (HCl), sodium hydroxide (NaOH) and sodium arsenate heptahydrate ($Na_2HAsO_4 \cdot 7H_2O$) (Sigma-Aldrich, Balestier Point, Singapore). The all chemicals used are analytical grade and directly used without further purification.

2.2. The Synthesis of Mn-Fe LDH Composites

The Mn-Fe LDH is synthesized by using the co-precipitation method with modification on the interlayer anion (Otgonjargal *et al.*, 2012). A mixed solution of ca. 700 mL containing 0.1 mol of $MnCl_2 \cdot 4H_2O$ and 0.1 mol $FeCl_3 \cdot 6H_2O$ (i.e. of the molar ratio of Mn^{2+}/Fe^{3+} was 1) was added dropwise under intense magnetic stirring of 200 rpm into 1500 mL 2 M NaOH solution until the well-mixed solution, while heated with a temperature of $45 \pm 2^\circ C$ during this process.

After that, the obtained slurry was reheating at $85^\circ C \pm 2^\circ C$ for 2 hours under slow mixing of 50 rpm. The slurry was kept at room temperature for around 12 hours, then separated the top solution and bottom reliable by using a pipette volume. The solid products were separated by centrifugation and washed with deionized water several times until the pH reaches a constant of 8.5–9.0, and the conductivity is constant. The Mn-Fe LDH was formed were dried at $60^\circ C$ for 24 hours, then were crushed and sieved to the size of 100 mesh for further experiment as an adsorbent in adsorption experimental.

2.3. The Batch Adsorption Experiment

A preliminary experiment was conducted by using synthetic wastewater to investigate the

arsenic removal process by using LDHs. Synthetic wastewater was prepared by diluting a 100 mg/L of As (V) stock solution into deionized water. Despite the presence of the arsenic in ground water and wastewater quite lower, the initial arsenic concentration of ca. 50 mg/L was chosen in this experiment to known the adsorption capacity of adsorbent. The pH of the arsenic solution was adjusted by using 2 M HCl and NaOH solution. The equilibrium time (5–300 minutes), the effect of equilibrium pH (5–10), temperature adsorption (30–60°C), and stoichiometry calculation were investigated and discussed in the following section. The batch experiment procedures were done in duplicate samples, and the average values was taken to present the results data.

2.4. Analysis and Characterization

The structure morphology properties and chemical composition properties of the Mn-Fe LDH have been characterized using scanning electron microscopy (SEM-EDX, JSM-6390 LV, JEOL USA Inc, USA) and X-ray diffraction (XRD, D/MAX-B, Rigaku Co., Japan). The surface morphology were observed by scanning electron microscopy (SEM) with energy-dispersive X-ray spectroscopy (EDX), then also to confirmed the oxidation states of Mn and Fe in the LDH sample. The X-ray diffraction (XRD) measurement was performed on Rigaku D/MAX-B X-ray diffractometer by using Copper K-alpha ($\text{CuK}\alpha$) radiation. The operation voltage and current were kept at 40 kV and 100 mA, respectively.

3. Results and Discussion

Exposition and discussion of the results of the study were investigated and discussed in the following section.

3.1. Characterization of Mn-Fe LDH

The SEM-EDX and XRD analysis were used to investigate characterization of the Mn-Fe LDH resulted from co-precipitation process method.

SEM images of the Mn-Fe LDH in Figure 1a confirmed the formation of a round shape structure with a particle size around of 1 μm with the heterogeneous structure. However, the aggregation of some particles was also observed. The heterogeneous structure promotes the adsorption density efficiency through the diffusion of the adsorbate onto the surface of the adsorbent.

As shown in Figure 1b SEM-EDX and Table 1, the ratio of metals is an essential parameter for characterizing LDH compounds. Mn's molar ratio to Fe was around 1.05 ($27.00/25.66 = 1.05$), where does it agree with the initial ratio of Mn/Fe (Otgonjargal *et al.*, 2012).

The XRD pattern shows sharp and symmetric peaks at lower 2θ values, which are characteristic of hydrotalcite-like compounds (lamellar compounds), and also indicate a high degree of crystallinity of the sample.

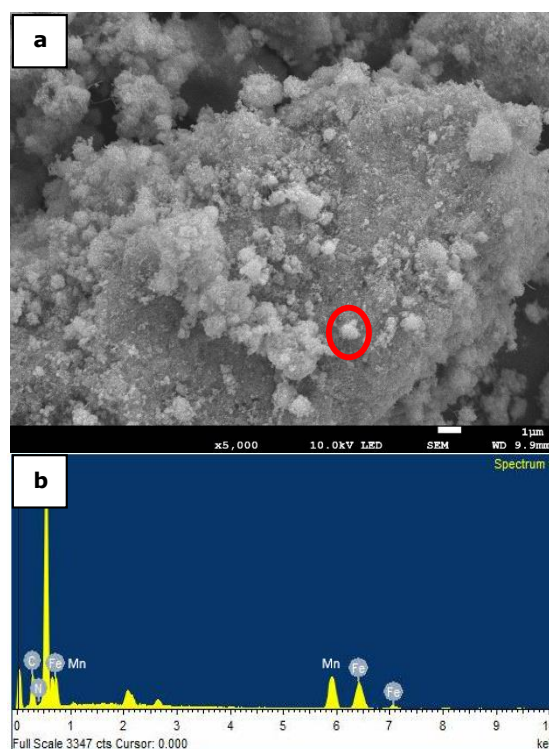


Figure 1. SEM image (a) and SEM-EDX analysis (b) of Mn-Fe LDH

Table 1. Chemical composition of Mn-Fe LDH by EDX

Chemical Element	Weight, %	Atomic, %
C	15.89	24.33
N	1.25	1.32
O	30.20	56.89
Mn	27.00	9.01
Fe	25.66	8.45

XRD pattern of the Mn-Fe LDH were observed their peaks at around 16, 24, 27, 31, 35, 39, 52, 55 and 57° as shown in Figure 2. The Mn-Fe LDH had some XRD peak patterns that matching those of from iron chloride and manganese chloride (Figure 2), which indicates that the LDHs have some crystalline structure from a simple physical mixture of iron and manganese chloride. The material

synthesized based on the structure of manganese chloride. The structure of the Mn is substituted by Fe to give positively charged sheets. This positive charge is balanced by anions such as hydroxide and chlorinate. Therefore, the generated sample is a chlorinate-containing layered double hydroxide.

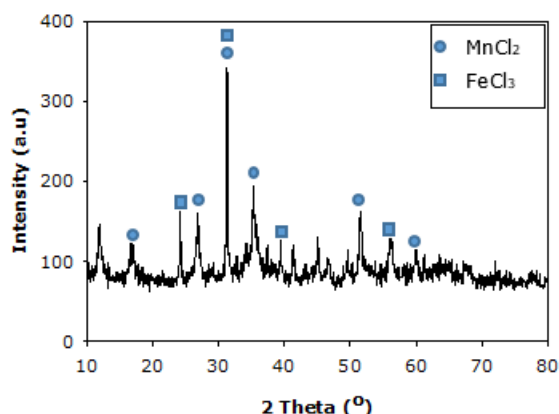


Figure 2. XRD analysis of Mn-Fe LDH.

3.2. Batch Adsorption Experiments

3.2.1. The Effect of Contact Time on As (V) Adsorption onto Mn-Fe LDH

Equilibrium time of arsenic removal process was determined by experimenting with room temperature, initial arsenate concentration of ca. 50 mg/L, LDHs dose of 0.5 g/L, pH_e 6.5–7.0, and various contact times from 5 to 300 minutes. Figure 3 shows that significant amount of arsenic was adsorbed in the first 10 minutes due to the concentration of solution and the vacant side of adsorption are quite high (Naswir *et al.*, 2020). After that, the removal of arsenic increased slowly until the equilibrium time was reached.

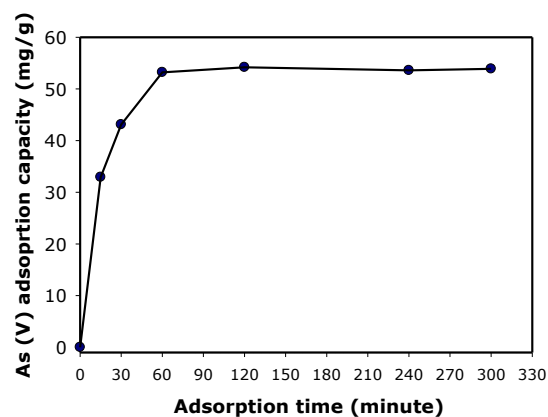


Figure 3. The effect of contact time on As(V) adsorption capacity at room temperature, stirring rates of 100 rpm, adsorbent dose of ca. 0.5 g/L, pH_e of 6.5 ± 0.5

The equilibrium time was reached the first time at 60 minutes. The arsenic adsorption on LDHs is a fast process, as reported in some previous studies by some researchers. The equilibrium time of the arsenic removal process by LDHs was around 30 hours and 2 hours written by Otgonjargal *et al.* (2012) and Suvokhiaw *et al.* (2016). However, contact time of 240 minutes (4 hours) was chosen for the further experiment to make sure that the system was already in equilibrium condition.

3.2.2. The Effect of equilibrium pH on As (V) Adsorption onto Mn-Fe LDH

In general, pH is considered an important parameter that controls the removal of contaminant from water and wastewater processes, either in the adsorption or ion exchange processes. The effect of equilibrium pH was studied by conducting experiment at room temperature, initial As (V) concentration of ca. 50 mg/L, LDHs dose of 0.5 g/L, the contact time of 4 hours, and over the equilibrium pH range 5 to 10. It is well known that the dissolution of LDHs takes place at low pH conditions. Thus, performing the removal process at pH higher than 7 will decrease the number of metals released from LDHs structure. However, the metal release from the LDHs structure depends on the type of metal present on the LDHs.

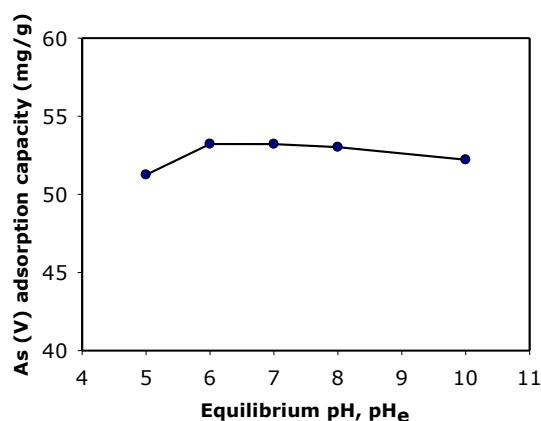


Figure 4. The effect of equilibrium pH (pH_e) on As (V) adsorption capacity at room temperature, stirring rates of 100 rpm, adsorbent dose of ca. 0.5 g/L, 4 hours.

Figure 4 shows As (V) uptake capacity increased with increasing pH and reached maximum at pH 6.5. But it was observed that the As (V) uptake capacity decreased with further increase in pH value up to 7.0. Therefore, the arsenate ion concentration in the solution is pH-dependent. In neutral conditions, As (V) species are completely in ionic form $H_2AsO_4^-$ and $HAsO_4^{2-}$. At pH below

the value of 3.0, the concentrations of the H_3AsO_4 are present. When the pH value increases (~ 11.0), AsO_4^{3-} will be the solution's dominant species. Arsenate was removed in the ionic form (H_2AsO_4^- and HAsO_4^{2-}) when ion exchange and adsorption process were the dominant mechanism. So, increasing pH value will increase arsenate uptake capacity due to the increase in arsenate ion species. However, a higher pH value will increase OH^- ions in the solution that will compete with arsenate ion for the LDHs surface site (Caporale *et al.*, 2011, Liu *et al.*, 2019, Otgonjargal *et al.*, 2012, Suvokhiaw *et al.*, 2016, Zubair *et al.*, 2017).

At a high pH value, the positive surface charge of LDHs will decrease. This might cause a reduction in an electrostatic attraction between anions in the solution and LDHs surface. There are some papers discussing the pH effect on arsenate removal by using LDHs. Some published reports indicated the removal of arsenate by LDHs was independent of pH, because LDHs have a high pH buffering capacity. Optimum pH for arsenate removal by using Fe_3O_4 was around 6.0 (Liu *et al.*, 2015). Similar results were obtained by Caporale *et al.* (2011), who investigated arsenate removal by using Mg-Fe LDH. Moreover, the lowest molecular interaction and these interactions appear because molecules have an unsymmetrical electronic distribution that creates small dipoles that can induce complementary dipoles in the neighboring surface, depending on the molecule's orientation respect the surface of the adsorbent. Furthermore chloride anion intercalated in the Mn-Fe LDH layers was exchanged with As (V). Kentjono *et al.*, 2010 was also reported that adsorption on LDHs was independent of the initial pH of the solutions because of high buffering capacity of the LDHs. When the initial pH was low, the pH increased due to LDHs dissolution, and resulting in the buffering of H^+ in the release of OH^- by LDHs. When the initial pH was high, the adsorption of OH^- from the solution by LDHs caused the pH decreased. Based on these above results the adsorption and ion exchange between interlayer chloride anions in hierarchical Mn-Fe LDH and As (V) solution was the main adsorption mechanism.

3.2.2. The Effect of Temperature on As (V) Adsorption onto Mn-Fe LDH

The temperature effect was studied to better understand the feasibility, spontaneity, and nature of the arsenate removal process. Experiments were carried out by varying

temperature from room temperature until 60°C at initial As (V) concentration of ca. 50 mg/L, pH_e of 6.5-7.0 and contact time of 4 hours. From Figures 5 we can see that the process was independent of temperature, since the curves were close to each other. Thus, LDHs can be applied in arsenate removal processes, either in low temperature or high-temperature processes. This would benefit wastewater treatment processes operate at low temperature (Iryani *et al.*, 2019, Kentjono *et al.*, 2010), since no need for temperature control during the treatment processes.

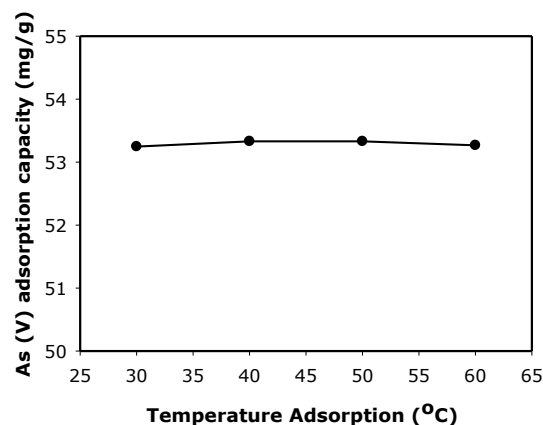


Figure 5. The effect of temperature on As (V) adsorption capacity at pH_e of 6.5 ± 0.5 , stirring rates of 100 rpm, adsorbent dose of ca. 0.5 g/L, 4 hours.

3.2.3. Adsorption isotherms studies

The adsorption isotherm was assessed by the Langmuir isotherm. Langmuir was developed assuming that a fixed number of accessible sites are available on the adsorbent surface, monolayer adsorption occurs, which its adsorption occurs when molecules adsorbed on the surface of adsorbent form a saturated layer. The equation of Langmuir is represented as follows (Langmuir, 1918).

$$q_e = \frac{K_L \cdot q_{\max} \cdot C_e}{1 + K_L \cdot C_e} \quad (1)$$

Where, q_e is the equilibrium As (V) concentration on the adsorbent (mol g^{-1}); C_e , the equilibrium As (V) concentration in solution (mol L^{-1}); $q_{\max}$, the monolayer capacity of the adsorbent (mol g^{-1}); K_L , the Langmuir constant.

The adsorption studies were conducted by varying initial As (V) concentration of ca. 5 to 100 mg/L at room temperature, pH_e of 6.5 ± 0.5 , adsorbent dose of 0.5 g/L, shaking rate of 100 rpm, and reaction time of 4 hours.

Figure 6 shown that As (V) adsorption uptake increased up to 50.21 mg/g. The Langmuir model described the adsorption data satisfactorily (R^2 value average = 0.970), and the batch maximum adsorption capacities was found as 63.09 mg/g at pH value of 6.5 ± 0.5 stirring rates of 100 rpm, adsorbent dose of ca. 0.5 g/L, 4 hours.

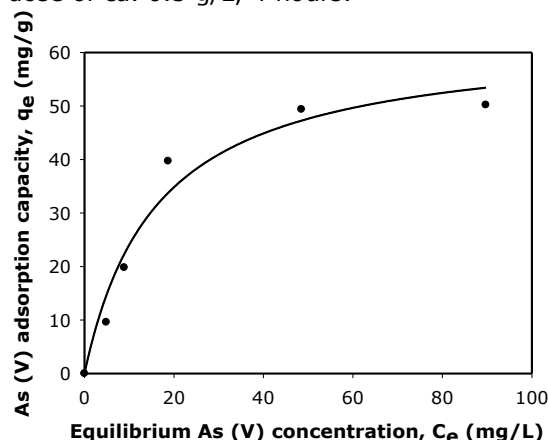


Figure 6. Adsorption isotherms of arsenate on the Mn-Fe LDH at pH value of 6.5 ± 0.5 stirring rates of 100 rpm, adsorbent dose of ca. 0.5 g/L, 4 hours.

Furthermore, this result was also confirm that chloride anion interlayer in the Mn-Fe LDH exhibit better As (V) adsorption uptake capacity than previous research using carbonate as interlayer (Otgonjargal et al., 2012).

Conclusions

Mn-Fe (Cl_2) LDH with a molar ratio of 1 : 1 of Mn/Fe was well synthesized using the co-precipitation method. SEM analysis results obtained round shape structure with a particle size around of 1 μm with the heterogeneous structure of Mn-Fe LDH structure in which this structure will be good for metal ions can diffuse and penetrate into the inside of each adsorbent particle and trapped into the LDH. The SEM-EDX measurement results from the Mn-Fe LDH ratio of 1.05 indicate that the Mn element is similarly dominant with the Fe element in the sample. Mn-Fe LDH showed up high efficiency in removing arsenate from synthetic acid mine drainage. The optimum pH for the arsenate removal process was around 6.5 ± 0.5 . The method of As(V) adsorption by LDH with a ratio of Mn-Fe LDH 1 : 1 is the promising adsorbent because it adsorption capacity to capture of arsenate anion to around 63.09 mg/g within an

equilibrium time of 4 hours, pH_e of 6.5 ± 0.5 , stirring rate of 100 rpm, and room temperature. The results demonstrated that Mn-Fe LDH is a potential adsorbent for arsenate removal and expands the application of LDH technology. Therefore, the Mn-Fe LDH has the potential to be used as an adsorbent in water and wastewater treatment.

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