Research Article

Study on Leaching of Phosphate from Municipal Wastewater Treatment Plant's Sewage Sludge and Followed by Adsorption on Mg-Al Layered Double Hydroxide

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Sewage sludge from municipal wastewater treatment plant was a rich phosphorus resource. In this study, HCl, H_2SO_4, and HNO_3 were investigated as leaching acids for extraction of phosphate from the sludge to recover this value nutrient by adsorption using layered double hydroxide (LDH) nanomaterial. Mg-Al LDH was synthesized by coprecipitation at room temperature and at a constant pH of 10 ($\pm$0.5) with Mg/Al molar ratio of 2.0. The material was characterized by scanning electron microscopy (SEM), X-ray diffraction (XRD), Brunauer-Emmet-Teller (BET), and Fourier transform infrared spectroscopy (FTIR) and then was used to adsorb phosphate leaching from the sewage sludge. Adsorption experiments were carried out as a function of adsorbent dose, phosphate initial concentrations, and pH. The adsorption isotherm data fitted the Langmuir model perfectly. The high adsorption capacity of Mg-Al LDH and slowly released phosphate of the post-adsorption LDH suggested that this material was an excellent adsorbent for phosphate recovery and could be considered as a potential phosphate release fertilizer.

1. Introduction

Phosphorus (P) is an essential nutrient for plant growth. In the form of phosphate, it was used to produce fertilizers in order to increase yields in agriculture. It also presents in many products that are widely used such as detergents, hardeners, toothpaste, corrosion inhibitors, industrial food additives [1]. However, the over-use of fertilizers, as well as phosphate-containing products causes a large amount of phosphate to accumulate in the soil or be washed out into rivers, streams, ponds, lakes, and present in wastewater contributes to significant phosphate pollution [2]. Pollution of phosphate in the aquatic environment can lead to the phenomenon called eutrophication, in which algal blooms excessively, dissolved oxygen will be largely consumed, so it causes serious effects on the quality of water and aquatic life [3].

In the face of such phosphate pollution, the treatment and control of phosphates in wastewater is a big challenge. According to data of the Vietnam Water Supply and Sewerage Association by the end of 2016, out of 35 centralized wastewater treatment systems operating in the country, only 7 biological treatment plants combine nitrogen and phosphate treatment thoroughly [4]. Nowadays, phosphate control is interested with the main research focusing on treatment techniques such as precipitation, adsorption, ion exchange, and biological treatment [4, 5]. However, we should know that the natural supplies of phosphate are nonrenewable and will be exhausted quickly. Because of the irreplaceable role of phosphate in fertilizer production, the objective of phosphate treatment should change from removal to recovery.

In conventional municipal wastewater treatment, it was reported that around 90% of incoming phosphorus load is concentrated in the sewage sludge [6]. Although sewage sludge from municipal wastewater treatment plant (MWTP) has high amounts of phosphate, it should not be used directly as fertilizer due to concerns about its consisting of heavy metals, pathogens, and other toxic matters [7].
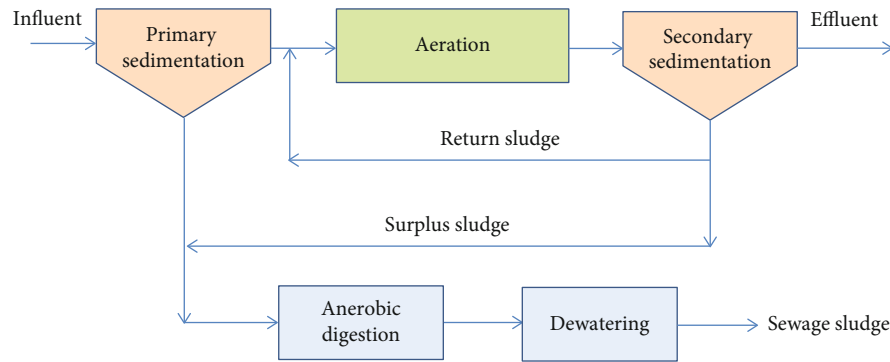
FIGURE 1: Municipal wastewater treatment system of Truc Bach plant.

Moreover, phosphate fix in the sludge exhibits low availability for plant to absorb [8]. So, the main technologies for phosphate recovery from sludge are leaching phosphate bound in the sludge into the liquid phase and then recovering phosphate from the supernatant by chemical precipitation or adsorption.

In order to extract phosphate from the sludge phase, wet chemical and thermochemical treatments are used [9]. In the thermochemical process, high purity of recovered phosphate is achieved but the high consumption of energy and specific equipment are needed [10]. In the wet chemical approach, both strong acids and alkalis were suggested. However, adding acids is reported more effective than adding alkalis. Alkali leaching achieves about 60-70% of total P separated from sewage sludge [11], while acid leaching can release over 80% of P [12].

After being released through the wet chemical process, the more effective method to recover phosphate in the leachate is adsorption. Recently, layered double hydroxides (LDHs), also known as hydrotalcite-like compounds or anionic clay, are considered promising materials that can effectively adsorb phosphates due to structural resemblance with the cation clays and exhibit high anion exchange capacity and good affinity for phosphate [13, 14]. LDHs are 2D nanomaterials that have the general formula $[M_{1-x}^{2+}M_x^{3+}(OH)_2]^{x+}[(A^{n-})_{x/n}.mH_2O]^{x-}$, where M^{2+} and M^{3+} are di- and trivalent metallic cations, respectively, and A^{n-} is an intercalated anion, which has the ability to be exchanged for other anions in solution giving special properties of LDHs [15]. LDHs have been proven as excellent adsorbents for phosphate removal from aqueous solutions in many research works [16].

It is indicated that phosphate ions are adsorbed on LDHs via electrostatic attraction, ligand exchange, and ion exchange [17, 18]. Previous studies mainly focused on phosphate recovery from waste streams by LDHs, followed by a release of phosphate from these materials by desorption solutions such as NaCl or NaOH [19]. Recently, it is noticed that LDHs are able to slowly release intercalated phosphate due to their unique structure [20]. After adsorption, the LDHs provide physical protection to phosphate, decreasing the direct contact of phosphate with the soil; therefore, phosphate can be released in a more controlled way. This means that phosphate loaded LDHs might be used directly as slow

TABLE 1: Total phosphorus and metal composition of the sewage sludge.

Composition	Concentration	Unit
Total P	3.4	%
Cd	0.839	μg/L
Mg	9.151	mg/L
As	0.071	mg/L
Sn	1.695	μg/L
Na	0.518	mg/L
Hg	7.382	μg/L
Zn	0.604	mg/L
Al	29.977	mg/L
Ca	14.376	mg/L
Mn	0.255	mg/L
Fe	11.360	mg/L
Ni	0.019	mg/L
Cr	0.045	mg/L
Cu	0.098	mg/L
Pb	0.032	mg/L

and controlled release P fertilizers in soils [21, 22]. The development of LDHs as fertilizers really makes sense to connect the recycling of phosphorus from waste streams and phosphorus fertilizers. Moreover, uptake of nutrients can be increased by encapsulating the phosphorus fertilizers in nanoform that provide gradual and controlled release of phosphate to the target sites through direct internalization of products, therefore ultimately reducing phosphorus loss and minimizing the risk of environmental pollution [23, 24].

In this paper, the wet chemical method using strong acids was tested for leaching of phosphate from municipal wastewater treatment plant's sewage sludge, and then recovery of phosphate in the leachate by adsorption onto Mg-Al layered double hydroxide nanomaterial was the aim of the study.

2. Materials and Methods

2.1. Sludge Characteristics. Sludge was obtained from Truc Bach MWTP in Hanoi, Vietnam. The wastewater treatment system of this plant is presented as in Figure 1.

TABLE 2: Equations and parameters of isotherm models.

Isotherm models	Equation	Parameters
Langmuir	$q_e = \frac{q_{max}bC_e}{1 + bC_e}$ $R_L = \frac{1}{1 + bC_0}$	q_e (mg/g) is the equilibrium adsorption capacity. q_{max} (mg/g) is the maximum adsorption capacity. b (L/mg) is the Langmuir isotherm constant related to the energy of adsorption. R_L is a factor of adsorption feasibility.
Freundlich	$q_e = k_f C_e^{1/n}$	k_f and n are Freundlich isotherm constants related to adsorption capacity and adsorption intensity.
Dubinin-Radushkevich	$q_e = q_m \exp\left(-\beta\varepsilon^2\right)$ $\varepsilon = RT\ln\left[1 + \frac{1}{C_e}\right]$ $E = \left[\frac{1}{\sqrt{-2\beta}}\right]$	q_m is the Dubinin-Radushkevich constant. β (mol²/kJ²) is the activity coefficient related to mean sorption energy. ε is the Polanyi potential. E (kJ/Mol) is the mean sorption energy.

The sewage sludge after dewatering was used in this study. Total phosphorus in the sludge was measured according to the procedure given in Vietnam Standards TCVN 8940-2011 [25]. Metals from the sludge were extracted in the ETHOS UP Microwave Digestion System according to U.S.EPA Method 3051A [26], and their concentration was measured on an inductively coupled plasma spectrometer (iCAP Q ICP-MS) according to SMEWW 3125:2017 [27]. The ICP measurements were made in duplicates to get the average data. Phosphorus and metal composition of the raw sewage sludge are presented in Table 1.

2.2. Acid Leaching of Sludge. The ability of leaching phosphate from the sludge was tested by the wet chemical method, which was individually carried out by using strong acids HCl, H_2SO_4, and HNO_3 at varying concentrations (0.05-2.0 mol/L HCl and H_2SO_4; 1.0-5.0 mol/L HNO_3). The liquid/solid (L/S) ratios of 10:1, 25:1, and 50:1 (mL/g) were examined. The leaching experiments were carried out at room temperature and shaken at 100 r/min for 24 h. After filtered through 1 μm ashless filter paper, the concentration of phosphate in the leachate was measured according to the Vanadomolipdophosphoric acid colorimetric method [28] and compared to its concentration in the raw sludge to calculate the leaching percentage; so, the optimal condition for leaching phosphate from the sludge would be determined.

2.3. Synthesis of Mg-Al Layered Double Hydroxide. In this study, Mg-Al LDH was synthesized by coprecipitation from the solution of $Mg(NO_3)_2 \cdot 6H_2O$ and $Al(NO_3)_3 \cdot 9H_2O$ (initial Mg/Al molar ratio of 2.0). The pH was maintained at 10 ± 0.5 by adding 2.0 M NaOH and 0.5 M K_2CO_3. The synthesis was performed at room temperature. The resulting precipitate was aged for 24 h and washed with deionized water until the effluent solution pH was neutral. After drying at 90°C overnight, the precipitate was lightly grounded to get particle size from 0.5 to 1.0 mm.

Crystal phases of the Mg-Al LDH were performed on Rigaku Miniflex 600 X-ray diffractometer using CuK-α radiation. Surface area and pore analysis were performed using the Brunauer-Emmet-Teller (BET) approach on a NOVA-touch LX4 adsorption analyzer. FTIR spectra were collected on a Jasco 4600 FTIR spectrophotometer. Scanning electron microscopy (SEM) was conducted to observe the surface
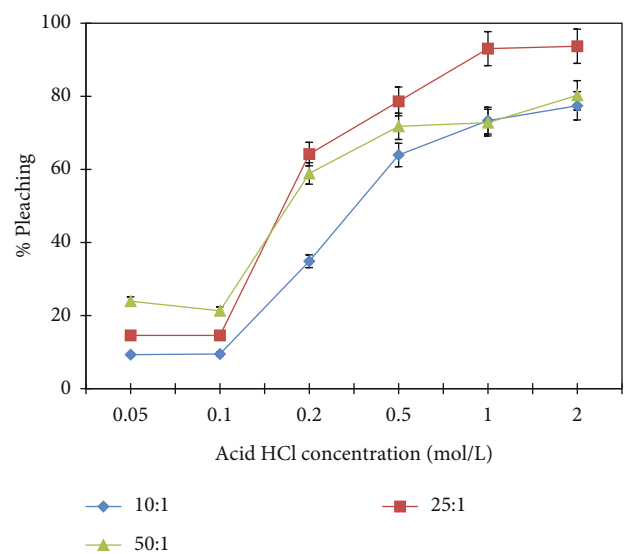

FIGURE 2: Effect of HCl concentration and L/S ratio on the leaching of phosphate from the sewage sludge.

morphological structure of synthesized Mg-Al LDH using a Hitachi S4800 scanning electron microscope.

2.4. Phosphate Recovery from Acid Leachate. Synthesized Mg-Al LDH was used to adsorb phosphate from acid leachate. The adsorption experiments were carried out in batches with 50 mL of adsorbed solution at neutral pH (adjusted by diluted NaOH) and at room temperature. After the equilibrium time (6 h), LDH was separated from the solution by centrifugation, and phosphate was analyzed in the solution by the Vanadomolipdophosphoric method. All the analyses were taken in triplicates. The adsorption capacity of the material q and adsorption efficiency E (%) was calculated according to the following formulas:

$$q = \frac{(C_0 - C_e)V}{m}, \tag{1}$$

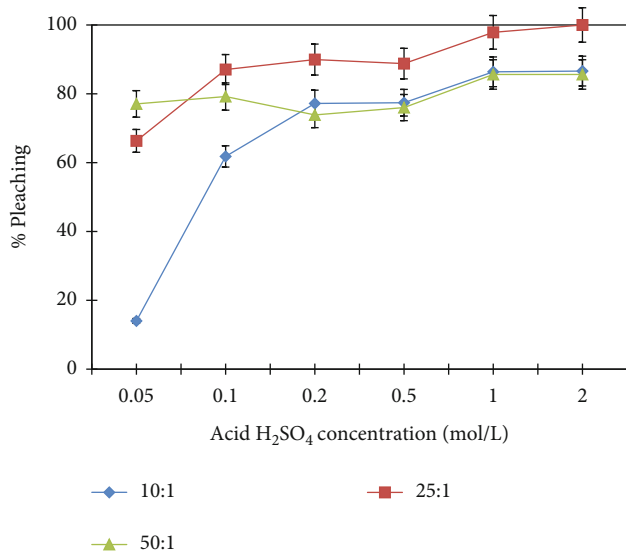$$E = \frac{(C_0 - C_e)}{C_0} \times 100\%, \tag{2}$$

FIGURE 3: Effect of H_2SO_4 concentration and L/S ratio on the leaching of phosphate from the sewage sludge.
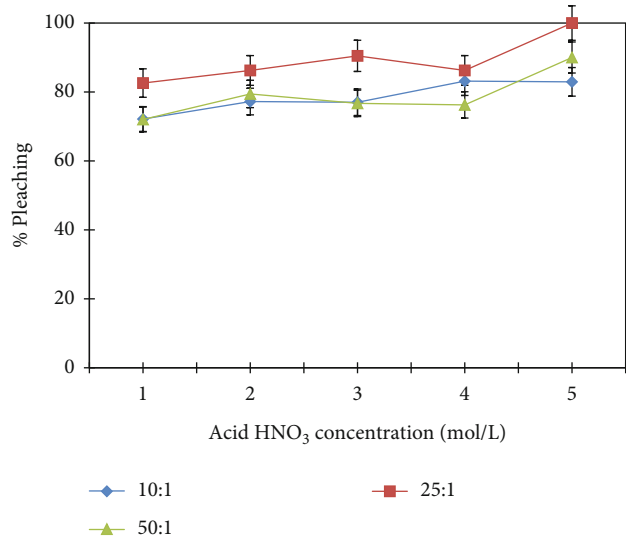

FIGURE 4: Effect of HNO_3 concentration and L/S ratio on the leaching of phosphate from the sewage sludge.

where C_0 and C_e are the initial and equilibrium concentration of phosphate in leachate (mg/L), q is the equilibrium adsorption capacity (mg/g), m is the adsorbent dry weight (g), and V is the adsorbed solution volume (L).

The influence of LDH dosage, the initial concentration of phosphate in leachate (adsorption isotherm), and pH are factors investigated in this work.

2.5. Adsorption Isotherms. The relationship between the amount adsorbed and adsorbate concentration in the aqueous phase at equilibrium was evaluated using three isotherm models: Langmuir, Freundlich, and Dubinin-Radushkevich models. Langmuir isotherm assumes the existence of a maximum adsorption limit corresponding to a complete monolayer formation of the adsorbate on the adsorbent surface.
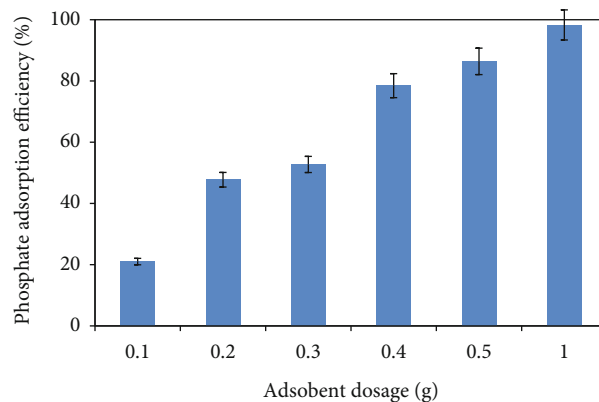

FIGURE 5: Phosphate recovery from acid leachate as a function of LDH dosage from 0.1 g to 1.0 g.
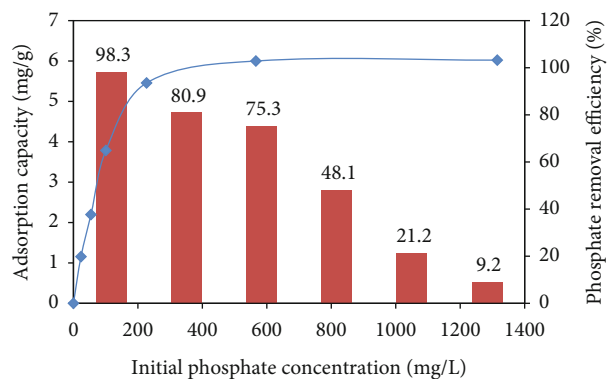

FIGURE 6: Adsorption capacity and removal efficiency of phosphate on Mg-Al LDH as a function of initial phosphate concentration.

The computed R_L value suggests the adsorption nature to be either unfavorable ($R_L > 1$), linear ($R_L = 1$), favorable ($0 < R_L < 1$), or irreversible ($R_L = 0$) [29]. The Freundlich model is often used to describe chemisorption on heterogeneous surfaces [30]. The Dubinin-Radushkevich model was commonly used to determine the adsorption type as physisorption or chemisorption [31]. Equations and parameters of the isotherm models are presented in Table 2.

2.6. Phosphate Release from LDH. To evaluate the applicability as a slow-release fertilizer of phosphate adsorbed LDH, 5 g of the material was put in a sealed plastic bottle containing 50 mL of the distilled water adjusted pH = 4, 6, and 8 at room temperature. After a certain time (1, 3, 5, 7, 10, 14, 28 days), the solution was decanted to determine the phosphate content, and another 50 mL of new pH adjusted water was added to the bottle to analyze for the next time. The solution was shaken well before taking samples for analysis.

3. Results and Discussion

3.1. Factors Affecting the Leaching Process. The effect of HCl, H_2SO_4, and HNO_3 on phosphate leaching from the sewage sludge at different concentrations and L/S ratios is depicted in Figures 2–4.

TABLE 3: Isotherm parameters for phosphate adsorption on Mg-Al LDH.

Freundlich isotherm		Langmuir isotherm		Dubinin-Radushkevich isotherm	
Parameters	Value	Parameters	Value	Parameters	Value
K_f (mg/g)	1.519	q_{max} (mg/g)	6.086	q_m (mg/g)	4.407
$1/n$	0.223	b (L/mg)	0.088	β (mol^2/kJ^2)	$1.00E-07$
R^2	0.931	R^2	0.999	R^2	0.671
		R_L	0.009-0.326	E (kJ/Mol)	1.581

The figures show that in most cases, the phosphate leaching increases with increasing acid concentration and gets the best results at the L/S ratio of 25 : 1. At this ratio, approximately 80% of phosphate was leached at a concentration of 0.1 mol/L and higher in the case of using H$_2$SO$_4$ and at higher than 0.5 mol/L and 1 mol/L in the cases of using HCl and HNO$_3$, respectively. H$_2$SO$_4$ seems to be the best acid for phosphate leaching. Considering the cost-effective consumption of acid and achievement of complete leaching of phosphate, 1.0 mol/L H$_2$SO$_4$ acid at the L/S ratio of 25 : 1 was chosen as the most suitable condition for phosphate leaching.

3.2. Recovery of Phosphate from Acid Leachate by Adsorption on Mg-Al LDH.

Phosphate from acid leachate was recovered by adsorption on the synthesized Mg-Al LDH. The phosphate adsorption was investigated at different adsorbent doses, phosphate initial concentrations, and pH values.

3.2.1. Effect of Adsorbent Dose.

The effect of Mg-Al LDH dosage from 0.1 to 1.0 g on the phosphate adsorption at the initial concentration of 23.46 mg/L (diluted acid leachate 100 times) was studied. As presented in Figure 5, the phosphate recovery increases with an increasing amount of the LDH. More than 80% of phosphate in leachate was uptaken at the adsorbent dose of 0.5 g, and nearly 100% was obtained at the dose of 1.0 g. So, the most suitable adsorbent dose for phosphate adsorption in 50 mL solution was 1.0 g.

3.2.2. Effect of Phosphate Initial Concentration.

Different initial phosphate concentrations were prepared by diluted the acid leachate at different ratios (1 : 100, 1 : 50, 1 : 25, 1 : 10, 1 : 5, 1 : 2).

The results illustrated in Figure 6 showed that when varying the initial concentrations from 23.5 to 1315.0 mg/L, the adsorption capacity firstly increased and then reached the equilibrium capacity. However, the percentage removal decreased from 98.3% to 9.2% with the increase in phosphate concentration. The decrease in phosphate adsorption efficiency could suggest the lack of an available area required for the high initial concentrations of phosphate.

Adsorption capacity at different equilibrium concentrations could be illustrated by the adsorption isotherm. The three commonly used isotherms, Langmuir, Freundlich, and Dubinin-Radushkevich isotherm models, were applied, and the summary data are presented in Table 3.

The results showed that the adsorption of phosphate on Mg-Al LDH was perfectly described by the Langmuir isotherm with $R^2 = 0.999$. The fitting of equilibrium adsorption
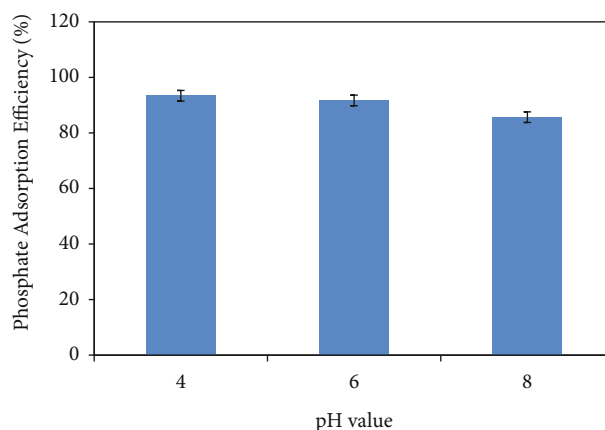

FIGURE 7: Efficiency of phosphate adsorption from acid leachate on Mg-Al LDH as a function of pH.
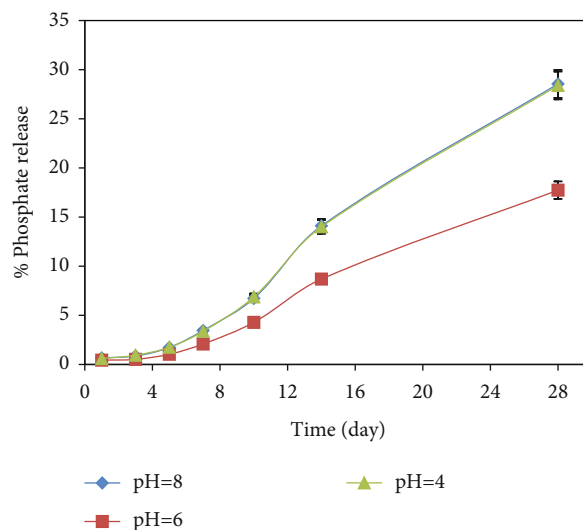

FIGURE 8: Efficiency of phosphate release from adsorbed Mg-Al LDH at pH = 4, 6, 8 using distilled water for desorption.

to Langmuir isotherm model suggests that monolayer and homogeneous adsorption onto the surface containing a finite number of identical sites were taken place [31]. This is in accordance with the anionic exchange mechanism involved in the phosphate adsorption on LDH. Moreover, the R_L values found in this study were between 0 and 1 which means the adsorption process is favorable. Furthermore, from the Freundlich isotherm data, the value of $1/n$ was

TABLE 4: Metal content in the adsorbed Mg-Al-LDH.

(a)

Metal	Cd	Pb	As	Cu
Content (mg/kg)	1.31	46.83	0.49	41.15
QCVN-01-189 : 2019/BNNPTNT	≤5 mg/kg	≤200 mg/kg	≤10 mg/kg	<1000 mg/kg

(b)

Metal	Fe	Mn	Zn	Ca
Content (mg/kg)	289.07	29.14	366.86	13,897.49 (or 1.38%)
QCVN-01-189 : 2019/BNNPTNT	<1000 mg/kg	<1000 mg/kg	<1000 mg/kg	<20%
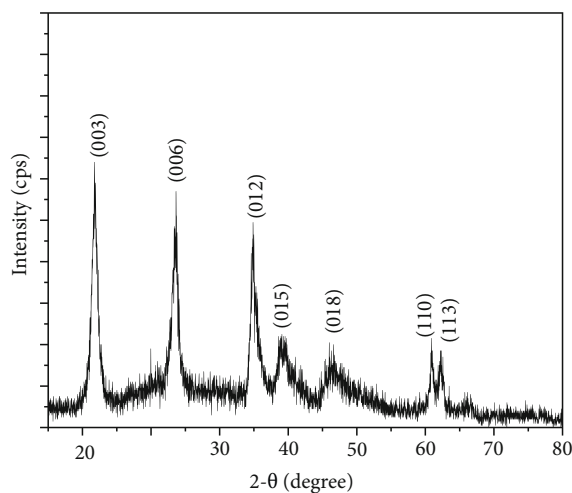
FIGURE 9: XRD pattern for synthesized Mg-Al LDH.

TABLE 5: Specific surface area, pore volume, and pore radius.

Sample	Specific surface area (m^2/g)	Pore volume (cm^3/g)	Pore radius (nm)
Mg-Al LDH BA	74.3603	0.19369	6.33512
Mg-Al LDH AA	36.0880	0.14390	7.97485
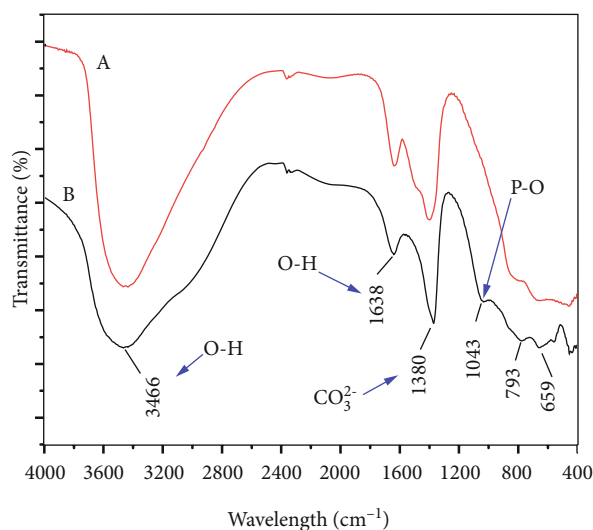
FIGURE 10: FT-IR spectra of Mg-Al LDH (a) before adsorption and (b) after adsorption.

found to be 0.2231 also indicating that the sorption of phosphate onto Mg-Al LDH is favorable. On the other hand, the mean sorption energy computed from the Dubinin-Radushkevich isotherm equation was determined to be 1.581 kJ/mol indicating a physisorption process. This means that besides the ion exchange mechanism, phosphate anion uptake on the LDH surface was also promoted by the electrostatic attraction between a surface positive charge and the anions.

3.2.3. Effect of pH Value. Phosphate adsorption was evaluated at three different pH values (pH = 4, 6, 8), which presented for the acid, neutral, and basic environment. The results presented in Figure 7 show that the adsorption efficiency decreased when the pH was higher. The adsorption taken at acid and neutral pH was more favorable than at basic pH. The reason for lower adsorption efficiency at higher pH was supposed due to competition with carbonate ions. Moreover, phosphate exists in the lower charge ($H_2PO_4^-$) at low pH that allows for higher adsorption [17].

3.3. Phosphate Released from Adsorbed Mg-Al LDH. After adsorption, phosphate-loaded Mg-Al LDH was examined by the phosphate release efficiency in water at three different pH values as shown in Figure 8.

After 28 days, the phosphate was released only 28.6%, 17.7%, and 28.4% at pH 8, 6, and 4, respectively. The release of phosphate by LDH was clearly slow, despite the process being incomplete, and it could be confirmed that the phosphate-loaded LDH can act as a slow-release fertilizer.

3.4. Evaluation of Metal Content in Adsorbed Mg-Al LDH. It is known that the raw sewage sludge contains metal components in certain concentrations. In the phosphate leaching process from the sewage sludge, strong acid is used, so it is inevitable that the metals will be dissolved into the leachate. To evaluate the possibility of metals being adsorbed on Mg-Al LDH during phosphate recovery from the leachate, the
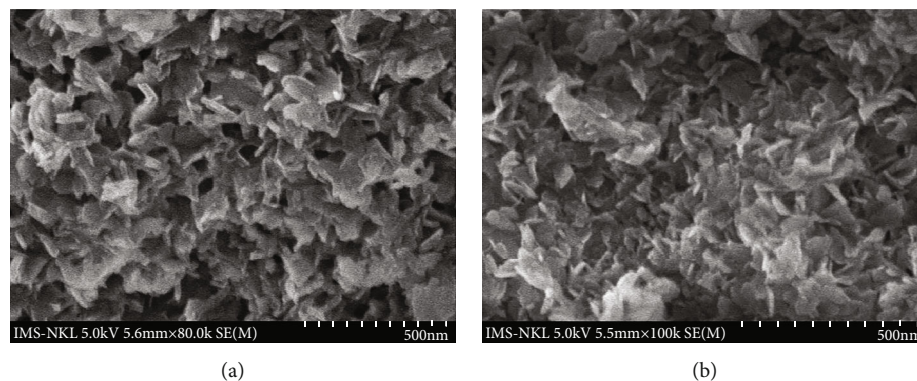
(a) (b)

FIGURE 11: Scanning electron microscopy images of the synthesized Mg-Al LDH (a) before adsorption and (b) after adsorption.

metal composition in the postadsorption Mg-Al LDH was analyzed according to SMEWW 3125:2017 [27].

For the purpose of using the postadsorption Mg-Al LDH as fertilizer, the results of metal composition in the material are compared to Vietnam National Technical Regulation on Fertilizer Quality QCVN-01-189:2019/BNNPTNT [32] as presented in Table 4. The results show that all metals, especially harmful metals such as Cd, Pb, As, and Cu, are much lower than the allowable limit. The presence of Fe, Mn, Zn, and Ca is partly beneficial because they are essential micronutrients for plants.

3.5. Characterization of Synthesized Mg-Al LDH.
XRD pattern of synthesized Mg-Al LDH presented in Figure 9 showed typical signal peaks for Mg-Al layered double hydroxide material at 2θ angles of 11.70°, 23.60°, 34.79°, 39.06°, and 60.92°, which are assigned to the crystal planes of (003), (006), (009), (007), and (110).

Corresponding to the (003) and (006) reflections, peaks with large intensity appear, showing the large distance between the interlayers. The basal spacing of (003) reflection of the material was found to be 7.6 Å, which was consistent with the previous reports [20, 33]. This means that the LDH material was successfully synthesized by the coprecipitation method.

BET surface area of Mg-Al LDHs before and after adsorption determined via the Brunauer-Emmett-Teller plot is presented in Table 5. Specific surface area of Mg-Al LDH before adsorption (BA) was similar to the Mg-Al hydrotalcite product of Sigma-Aldrich ($78 m^2/g$) [33] and higher than typical values of the specific surface area measured by BET range from 24.6 to 69 m^2/g of this material [34]. After adsorption, the occupation of phosphate on the surface caused a decrease in pore volume, a slight increase in pore size and a considerable decrease in specific surface area.

Mg-Al LDH materials before and after phosphate adsorption were also analyzed by infrared spectrum. The results are shown in Figure 10.

On the FTIR spectrum of the LDH before and after adsorption, spectral lines typical for Mg-Al layered double hydroxide can be seen. The broad absorption band in the range 3800-2500 cm^{-1} is attributed to the valence vibrations of the O-H group in the LDH molecule and of the interlayer adsorbed water molecules. The peak located at 1638 cm^{-1} is

assigned to the O-H bending vibration and indicated the presence of interstitial water molecules. Other absorption bands in the region below 1000 cm^{-1} characterize the oscillations of Mg-O and Al-O bonds in LDH. The new peak appearing at 1043 cm^{-1} on the postadsorption material is believed to be of the P-O bond, which gives information about the inner spherical surface (M-O-P) complex between phosphate and LDH [35]. It is therefore suggested that the surface hydroxyl groups (M-OH) of LDH are likely to be exchanged by phosphate ion, which is reported as the main mechanism for phosphate adsorption on LDH material [20].

The morphology of synthesized LDHs before and after adsorption is provided in Figure 11. SEM images show a well-developed layered structure with a fine dispersion of the platelet-like particles having estimated lateral size around a few tens of nanometers, and thickness is about a few nanometers. It also reveals relatively obvious hexagon overlapping crystals, which are typical for LDHs. The morphology features do not change after the adsorption and are similar to those observed in previous studies [14, 30, 36].

High specific surface area, unique morphology, and nanoparticle size are extremely important properties that make LDH have excellent phosphate adsorption capacity and well-controlled release ability.

4. Conclusions

The study of phosphate extraction from municipal wastewater treatment plant's sewage sludge has been performed for the view of recovery phosphate by Mg-Al layered double hydroxide. It could be confirmed that sewage sludge from municipal wastewater treatment plant was a beneficial source to recover phosphate. The conditions for above 80% of phosphate leaching were 0.1 mol/L H_2SO_4 or 1 mol/L HCl or 1 mol/L HNO_3 at the L/S ratio of 25:1 for 24 hours shaking at room temperature (~30°C). Mg-Al LDH with Mg:Al molar ratio of 2:1 was successfully synthesized and applied to adsorb phosphate from acid leachate. The synthesized LDH as a nanomaterial showed excellent phosphate adsorption efficiency at experimental conditions. Phosphate uptake by Mg-Al LDH fitted the Langmuir adsorption isotherm model perfectly and was intercalated in the LDH by exchange with ion carbonate. The LDH after adsorption could slowly release phosphate, which appears to be a

promising controlled release phosphate fertilizer. Metal content, especially harmful heavy metals such as Cd, Pb, As, and Cu in the adsorbed LDH, is much lower than the maximum allowable content specified in Vietnam National Technical Regulation on fertilizer quality. Thus, phosphate recovery from MWTP's sewage sludge by Mg-Al LDH after acid leaching may contribute to solving the problem of phosphate deficiency.

Data Availability

Data are available on request.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

Acknowledgments

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