Utilization of Lanthanum Carbonate and Bentonite for Phosphorus Removal from Domestic Sewage Effluent

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Abstract: lanthanum carbonate and bentonite were used as adsorbents to remove phosphorus from domestic sewage effluent. Three batch-mode adsorption experiments (using $La_2(CO_3)_3$, Bentonite/$La_2(CO_3)_3$ and Bentonite+$La_2(CO_3)_3$ as adsorbents) were carried out to investigate the effect of adsorbents for the removal of phosphorus. The experimental results show that Bentonite/$La_2(CO_3)_3$ and Bentonite+$La_2(CO_3)_3$ of 320mg/L can reduce phosphate concentration down to 0.06 mg P/L and 0.04 mg P/L from an initial value of 0.609 mg P/L at the contact time of 48 h, respectively.

1 INTRODUCTION

Excess phosphorus (P) in freshwater bodies is one of the major causes of eutrophication (Zhu et al., 2016). The regulations for removing phosphate from water/wastewater treatment applications are becoming more stringent (Zhao et al., 2014). Therefore, phosphorus removal from water/wastewater has attracted considerable research interest in the last few decades (Zhang et al., 2011). And a range of methods have been developed, mainly including biological, chemical, and physical treatments. Among these methods, the adsorption process is promising for phosphate removal, attributed to its attractive advantages of simple operation, high removal efficiency and fast adsorption rate, especially at low phosphate concentration (Mor et al., 2016). Many inorganic and organic adsorbents as well as industrial by-products, and biological wastes have been used in the treatment of phosphorus in water/wastewater.

Lanthanum-based adsorbents are gaining attention for the adsorption of phosphorus from water/wastewater in terms of their high affinity for phosphate and the lanthanum–phosphate complex forms, even when present in low concentrations of phosphorus. Various salts of lanthanum, such as $La_2(SO_4)_3$, $La(NO_3)_3$, $La(OH)_3$ and La_2O_3, have attracted intensive research interest in practical applications (Zhang et al., 2011; Yang et al., 2011; Zhang et al., 2016). However, some typical problems still exist which limit the wide application of these adsorbents. For example, it is found that most of their adsorption capacity is greatly declined in neutral and basic conditions. And the incomplete accessibility of La to phosphate in bulk samples is considered to be another reason for the reduced efficiency of La removal. Benefiting from the low leaching of lanthanum carbonate during phosphate adsorption, lanthanum carbonate can exhibit much higher safety and environmental friendly characteristics. Moreover, the pH buffering of carbonate salts is helpful for lanthanum carbonate show the observed small increase in pH value. Unfortunately, lanthanum carbonate is mainly used in the treatment of hyperphosphatemia of chronic kidney disease (Aaseth et al., 2018), very limited work has been done for dephosphorization agent outside the medical field.

The object of present paper is to provide lanthanum carbonate and bentonite for the treatment of phosphorus in domestic sewage effluent. Experimental results clearly indicate that the combined effect of lanthanum carbonate and bentonite significantly improved the removal efficiency of phosphorus in domestic sewage effluent.

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2 MATERIALS AND METHODS

2.1 Materials

All the chemicals used in this study were of analytical grade and were used without further purification. Lanthanum chloride hexahydrate ($LaCl_3·6H_2O$) was provided by Desheng New Material Co., Ltd. (Shandong, China). Sodium bicarbonate ($NaHCO_3$) and Sodium bicarbonate heptahydrate ($MgSO_4·7H_2O$) were obtained from Zhanyun Chemical Co., Ltd. (Shanghai, China). Sodium hydroxide ($NaOH$) and potassium dihydrogen phosphate (KH_2PO_4), were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Bentonite was supplied by Zhejiang Fenghong New Material Co. Ltd, Zhejiang, China.

2.2 Adsorbate Solution

The phosphorus solutions were collected from rural domestic sewage effluent in Zhuji, Zhejiang province, China. And the initial total P concentrations was 0.609 mg P/L.

2.3 X-Ray Diffraction (XRD) Analysis

The prepared lanthanum carbonate was analyzed using different equipments to reveal their physical properties. And the crystal structure of the adsorbents was analyzed using Bruker D8 Advance diffractometer (X'Pert Powder, PANalytical, Netherlands) with Cu-Ka radiation (40 kV, 40 mA) over the 2θ range of 10-60°.

2.4 Preparation of Lanthanum Carbonate

Lanthanum carbonate was synthesized via conventional coprecipitation method. The Mg^{2+} solution ($MgSO_4$) was added into the 0.4M La^{3+} solution ($LaCl_3$) at room temperature. Then 1.2M $NaHCO_3$ was added droplet under stirring and allowed to react for another 2hr. After the reaction mixture was stood for additional 1 hours, the mixed compounds were filtered and subsequently washed thoroughly with ethanol and deionized water until the pH of the effluent solution was neutral. Finally, the washed samples were oven-dried at 65°C for 12h to obtain the corresponding lanthanum carbonate adsorbents.

2.5 Batch Experiments

All batch adsorption experiments were performed in duplicate at room temperature using 25 ml glass tubes. Various concentrations (20-320 ppm) of adsorbents were introduced into 25 ml of adsorbate solution with an initial total P concentration of 0.609 mg P/L and a pH∼7. The mixing of adsorbents and adsorbate solution was performed by resting there for specified time. After that, the suspension was immediately centrifuged for phase separation. The supernatant was collected through filtration by using 0.45μm syringe filter, and analyzed to determine the residual P concentration by Mo-Sb anti-spectrophotometer method using a UV-Vis spectrophotometer (HACH, DR900, America).

The removal rate (R) of P was calculated from the following relation:

$$R = \frac{(C_0 - C_e)}{C_0} \times 100 \tag{1}$$

In the above equation C_0 represents initial concentration whereas C_e as final level of phosphate (mg P/L) in aqueous solution.

3 RESULTS AND DISCUSSION

3.1 The X-Ray Diffraction Analysis of Lanthanum Carbonate Adsorbents

The structure of lanthanum carbonate adsorbents was confirmed by the XRD analysis. As seen from Figure 1, the XRD patterns of the as-synthesized lanthanum carbonate ($La_2(CO_3)_3$) are typical structure of lanthanum carbonate with sharp peak at 10.27° corresponding to the (002) plane, which are highly crystalline structure of $La_2(CO_3)_3·8H_2O$ (PDF #25-1400), and broad peaks located at 18.51°, 20.79°, 21.24°, 27.08°, 29.06°, corresponding to the crystal surfaces of $La_2(CO_3)_3·8H_2O$ (020), (004), (022), (220) and (222) planes.

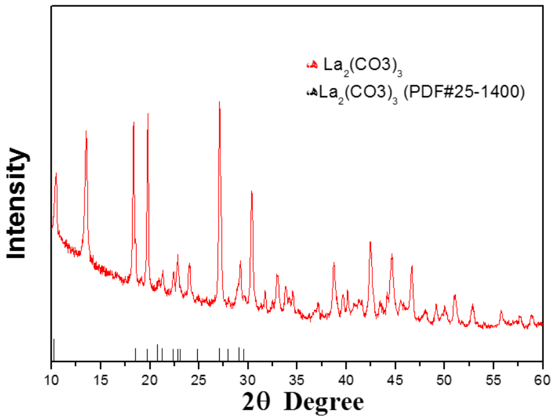

Figure 1: X-ray diffraction analysis of lanthanum carbonate.

3.2 Adsorption Experiments

Adsorption experiments were conducted using the synthesized adsorbents to evaluate their affinity towards phosphorus. In batch experiments, they were classified into three main categories: batch experiments I using only lanthanum carbonate as adsorbent, denoted as $La_2(CO_3)_3$; batch experiments II using the mixture of lanthanum carbonate and bentonite as adsorbent which is Bentonite/$La_2(CO_3)_3$; and batch III are batch experiments which add bentonite first, then followed by $La_2(CO_3)_3$, denoted as Bentonite+$La_2(CO_3)_3$. Their results for phosphorus removal are shown in Figure 2. It can be seen that as the adsorbents dose increases, percentage of phosphorus removal increased in three batch experiments. This might be due to the increased surface area of the adsorbent with increased adsorbent dosage. Further, it is noticed that the maximum removal of phosphorus, which was achieved using Bentonite/$La_2(CO_3)_3$ or Bentonite+$La_2(CO_3)_3$ as adsorbents, are 89.6% and 93.6%, respectively. These high phosphorus removal rates indicate that addition of bentonite can promote the adsorption of phosphate because of the more positive charges on the adsorbents. In this way, after phosphorus component being efficiently adsorbed by nano-prepared lanthanum carbonate particles, effect of flocculation by bentonite on nano-products made the continued separation process efficient and complete. Then, the improved phosphorus removal can be obtained from domestic sewage effluent. Meanwhile, it is found that feeding method also affects the results of phosphorus removal. As compare to batch experiments II using the mixture of lanthanum carbonate and bentonite as adsorbent, batch experiments III shows higher phosphorus removal ability. This might be attributed to the flocculation pretreatment of bentonite in batch experiments III. So that lanthanum carbonate can dephosphorize in a cleaner environment and exhibit better treatment effect.

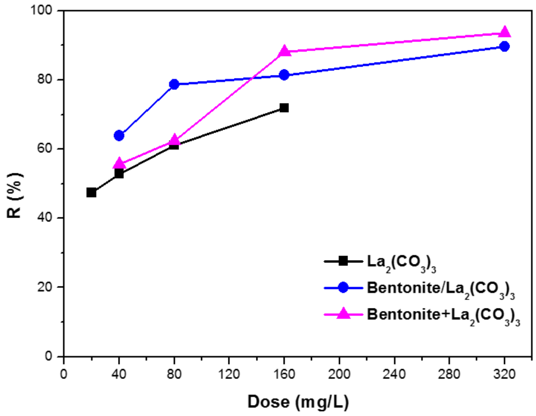

Figure 2: Effect of different types of adsorbents on phosphorus removal efficiency.

3.3 Effect of Contact Time

In order to investigate the effect of adsorption and desorption equilibrium on phosphorus treatment, results of phosphorus removal were investigated at the contact time of 24hr and 48hr, respectively. It can be seen in Figure 3 that when using $La_2(CO_3)_3$ alone as the adsorbent, phosphorus removal rates is positively correlated with the contact time for different in feed $La_2(CO_3)_3$ concentration. However, Inconsistent conclusions appeared for the other two adsorbents, that is, as the initial amount of bentonite and $La_2(CO_3)_3$ increase, phosphorus removal rate increases first and then decreases when Bentonite/$La_2(CO_3)_3$ and Bentonite+ $La_2(CO_3)_3$ were added into the solution. This phenomena indicates spontaneous nature of adsorption for phosphorus removal. When the equilibrium was achieved, no further increase in phosphorus adsorption was observed. Furthermore, it seems that phosphorus removal rate of Bentonite/$La_2(CO_3)_3$ and Bentonite+$La_2(CO_3)_3$ decreased as the contact time larger than equilibrium time. This may be due to the different stability between the two adsorbent components.

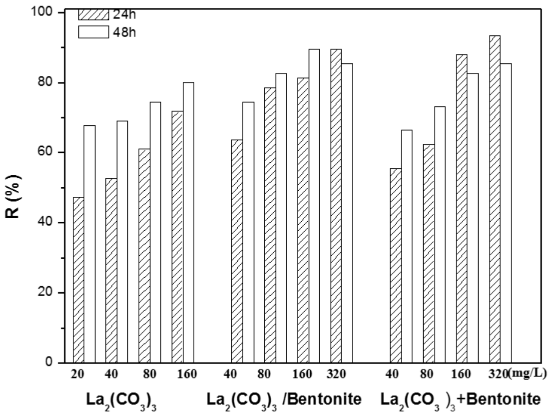

Figure 3: Adsorption of phosphorus with respect to the contact time.

3.4 Treatment Capacity

The correlation of treatment capacity and concentration is given in Table 1. It can be seen that the estimated effluent treatment capacity ranged 1.63-11.36 and 1.63–10.12 mg/g for Bentonite/$La_2(CO_3)_3$ and Bentonite+$La_2(CO_3)_3$, respectively, which clearly indicates their removal potential for phosphorus treatment. Owing to the repulsion between the negatively charged PO_4^{3-} species and negatively charged surface sites of bentonite, it is difficult for bentonite itself to adsorb phosphorus in domestic sewage effluent. Apart from the samples without bentonite, the best performing indicated the importance of cooperation between lanthanum carbonate and bentonite for phosphorus removal.

Table 1: Phosphorus treatment capacity at different adsorbents concentration.

Dose (mg/L)	Bentonite/$La_2(CO_3)_3$	Bentonite+$La_2(CO_3)_3$
40	11.36	10.12
80	6.30	5.58
160	3.41	3.15
320	1.63	1.63

4 CONCLUSIONS

The results of this study indicate that combination of lanthanum carbonate and bentonite is very good adsorbents for phosphorus disposal in domestic sewage effluent. The results obtained for the phosphorus removal were compared and the findings showed that Bentonite/$La_2(CO_3)_3$ and Bentonite+$La_2(CO_3)_3$ have good potential for removing phosphorus from domestic sewage effluent as compared to $La_2(CO_3)_3$. In this work, 89.6% and 93.6% removal of phosphorus were obtained by using Bentonite/$La_2(CO_3)_3$ and Bentonite+$La_2(CO_3)_3$ at the dose of 320 mg/L for 48hr in the resting experiment.

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