Applied Clay Science 156 (2018) 61–68

Contents lists available at ScienceDirect

Applied Clay Science

journal homepage: www.elsevier.com/locate/clay

Research paper

Ultrahigh-capacity and fast-rate removal of graphene oxide by calcined MgAl layered double hydroxide

Xiaoya Yuan*, Jiawei Niu, Yajie Lv, Qiuye Jing, Li Li

College of Materials Science and Engineering, Chongqing Jiaotong University, Chongqing 400074, China

ARTICLE INFO

Keywords:
Graphene oxide
Calcined MgAl-layered double hydroxide
Removal
Memory effect
Aggregation

ABSTRACT

Graphene oxide (GO) as an important derivative of graphene is potential toxic to cells and animals. In this study, MgAl-mixed metal oxide (MgAl-MMO) prepared by one-step calcination of MgAl-layered double hydroxide (MgAl-LDH) at 500 °C was employed to remove GO from aqueous solution. The effect of contact time, sorbent dosage, initial GO solution pH and co-existing anions on the removal capacity of MgAl-MMO composite was investigated in detail. The results showed that the sorbent dosage and the initial solution pH had significant effect on the adsorption process. MgAl-LDH composite exhibited ultrahigh GO adsorption capacity (984.2 mg/g at pH = 2) with low dosage (25 mg/L) and fast GO-eliminating rate (within 5 h). MgAl-MMO composite could be recycled by simply calcining the rehydrated production and reused for 5 times without significant loss of the removal capacity. This adsorption process could be well fitted by the pseudo-second-order model. The mechanism study further showed that the excellent removal performance of MgAl-MMO composite came from the memory effect of calcined LDH and pH-induced aggregation of GO.

1. Introduction

Graphene oxide (GO), as one of the most important graphene derivatives, has abundant hydroxyl and epoxy groups at basal plane, as well as carboxyl and carbonyl groups situated at the edges (Dreyer et al., 2010). Due to its intriguing structure and outstanding physicochemical properties, GO was considered to be a promising candidate in many multidisciplinary applications, such as sensors (Georgakilas et al., 2016), water treatment (Chabot et al., 2014), energy storage devices (Chen et al., 2012; Dai, 2013), catalysis (Xiang et al., 2012) and composites (Huang et al., 2012). Because of the hydrophilic oxygen-containing functional groups on the sheet surface, GO sheets are well dispersed in water. During the manufacture, application and disposing, GO is inevitably released into environmental and ecological system. Recent concerns about the fate and biological risks show that GO is potential toxic to cells and animals, including bacterial cells and humans. Duch et al. investigated graphene biocompatibility in the lung and found that GO caused severe and persistent lung injury due to chemical composition (Duch et al., 2011). GO could deactivate the *Escherichia coli* bacteria via damaging cell membrane because of the sharp edges of the GO nanowalls (Akhavan and Ghaderi, 2010) and caused cytotoxicity in human skin fibroblast cells and HeLa cells, in addition to dose-dependent hemolytic activity in human red blood cells (Liao et al., 2011; Zhang et al., 2012). Ahmed and Kryuchkova recently reported that GO significantly reduced the microbial metabolic activity (Ahmed and Rodrigues, 2013) and was highly toxic to one of the most common fresh water ciliate protest *P. caudatum* at the low concentration (Kryuchkova et al., 2016), respectively. So it is necessary to remove GO from natural aquatic environment.

Among various methods employed to remove aqueous organic and metallic pollutants, adsorption technique has been widely used because it is simple, economical, and cost-effective and a number of adsorbent materials have been studied extensively to remove organic dyes and heavy metal ions from aqueous solutions (Gupta and Suhas, 2009; Khan and Lo, 2016). Recently, some flocculants and adsorbents were developed for the removal and coagulation of GO. Wang's group found that the MgAl- and CaAl-LDH, Al_2O_3 particles, TiO_2 and chitosan-metal oxide composites could be potential coagulants for the elimination of GO from wastewater and the coagulation process was controlled by electrostatic interactions, hydrogen bonds and bridging function between oxygenated functional groups on GO sheets and the surface of the prepared samples (Ren et al., 2014; Yu et al., 2016; Wang et al., 2016, 2017a; Zou et al., 2016a, 2016b). Very recently, Wang et al. reported that La-doped MgAl- and CaAl-LDH exhibited rapid coagulation of GO with maximum capacity of 565.8, 558.6 mg/g, respectively and the coagulation of GO on La-doped LDH was mainly dominated by electrostatic attraction and hydrogen bond (Wang et al., 2017b). Unfortunately, most of the current materials used to eliminate aqueous GO

* Corresponding author.
E-mail address: yuanxy@cqjtu.edu.cn (X. Yuan).

https://doi.org/10.1016/j.clay.2018.01.018
Received 8 November 2017; Received in revised form 16 January 2018; Accepted 18 January 2018
0169-1317/ © 2018 Elsevier B.V. All rights reserved.

suffer low efficiency because the intrinsic coagulation mechanism involves physical process. Therefore, it is urgent to explore new materials with high removal capacity for the effective elimination of GO from aqueous solutions.

LDH are a class of lamellar two-dimensional anionic clays composing of positively charged brucite-like layers and exchangeable interlayer anions and their basic structure can be described by the general formula $[M^{2+}_{1-x}\ M^{3+}_{x}\ (OH)_2]^{x+}\cdot(A^{n-})_{x/n}\cdot mH_2O$, where M^{2+} and M^{3+} are di- and trivalent cations, respectively, and A^{n-} is the interlayer anions which balance the positive charges on the layers, x is defined as the molar ratio of $M^{2+}/(M^{2+} + M^{3+})$ (Evans and Slade, 2006; Mohapatra and Parida, 2016). Thermal treatment of LDH at moderate temperature usually leads to topotactic transformation to MMO (Zhao et al., 2010) and the resulting MMO material can restore the original LDH layered structure upon contact with aqueous solution and a variety of anions can be intercalated into the layer of the restored LDH in the rehydration process, which is known as the memory effect of MMO and thus makes MMO an attractive adsorbent for the removal of anionic pollutants from water (Das et al., 2003, 2006, 2007; Ni et al., 2007; Goh et al., 2008; Li et al., 2014; Yuan et al., 2013). The GO sheets are negatively charged as an anionic form due to the deprotonation of the carboxyl groups over a whole pH range (Yang et al., 2016). So from this point, GO could be inserted into the interlayer of the reconstructed LDH during MMO rehydration and concomitantly cleanup of GO from the solution is also achieved. In this study, MgAl-MMO with Mg/Al molar ratio of 3 was prepared by one-step calcination of MgAl-LDH at 500 °C and the effect of initial solution pH, adsorbent dosage, coexisting anions and contact time on its GO adsorption capacity was investigated in detail.

2. Experimental

2.1. Adsorption tests

All the reagents used and the detailed preparations of GO and MgAl-MMO composite were presented in the Supporting Materials. The batch adsorption experiments using MgAl-MMO were carried out in a 1000 mL conical flasks under 1000 rpm constant stirring with variation of initial solution pH, contact time, sorbent dosage and coexisting anions. All the tests were conducted under N_2 atmosphere unless stated otherwise. The solution pH was adjusted on a pHS-25 digital pH meter (INESA Instruments Ltd., Shanghai) by 0.01 mol/L aqueous HCl or NaOH solution. The competitive adsorption of GO in the presence of another anions was conducted by adding Na_2SO_4, Na_2CO_3, and NaCl (0.01 mol/L). At certain time intervals (0, 2, 4, 6, 8, 10, 20, 24, 28, 32, 36, 44, 52, 55 h), 5 mL of the solution aliquots was withdrawn, centrifuged at 10,000 rpm for 10 min and then the residual GO concentrations in the supernatant were analyzed by UV–Vis spectrophotometer at the wavelength of 227 nm. Each batch adsorption experiment lasted for about 55 h. The adsorption capacity of GO onto MgAl-MMO and the percentage of removal (% removal) were calculated as follows:

$$q_e = \frac{(c_0 - c_e)V}{m} \quad (1)$$

$$\%\text{removal} = \frac{C_0 - C_e}{C_0} \times 100\% \quad (2)$$

where q_e (mg/g) is the amount of GO adsorbed at equilibrium, c_0 and c_e (mg/L) are the initial and equilibrium concentrations of GO in the solution, respectively, m (g) is the mass of dry adsorbent used, V (L) is the initial volume of the GO solution. For recycle experiments, the solid was collected by centrifugation and washed with DI water. Then the regenerated MMO was obtained by calcining the dried solid at 500 °C for 4 h.

2.2. Characterization

Scanning electron microscopy (SEM) images were obtained with a Hitachi SU8020 scanning electron microscope with acceleration voltage of 20 kV. High-resolution transmission electron microscopy (HRTEM) were performed on a FEI Tecnai G2 F20 field-emission transmission electron microscopy at an accelerating voltage of 200 kV. The powder X-ray diffraction (XRD) patterns were recorded on a PANalytical X'pert Pro powder diffractometer with Cu-Ka radiation (λ = 1.5418 Å) with a scan step of 0.013°. Zeta potential was determined by ZetaSizer NanoZS (Malvern Instruments, Worcestershire, U.K.). X-ray photoelectron spectroscopy (XPS) measurements were performed on a Thermo Fisher ESCALAB 250Xi photoelectron spectrometer using monochromatic Al Kα X-ray source ($h\nu$ = 1486.6 eV).

3. Results and discussions

3.1. Removal of aqueous GO using MgAl-MMO sorbent

As presented in Fig. S1, the GO nanosheets are highly negatively charged from pH 2 to 10 due to the deprotonation into an anionic form and thus it could be incorporated into the interlayer of the rehydrated LDH due to the memory effect of the MMO. Before conducting the GO adsorption experiments, one must consider how to monitor the concentration of GO solution during the whole adsorption process. Lots of control experiments were tried and it was found that the adsorption intensity of aqueous GO solution at 227 nm obeys Beer-Lambert Law at GO concentration below 30 mg/L (Fig. S2) (Wang et al., 2016). To get the maximum adsorption capacity of MMO toward GO, an elaborate experiment was tried, where low sorbent dosage and high GO volume (1 L) were used to reach the sorption equilibrium.

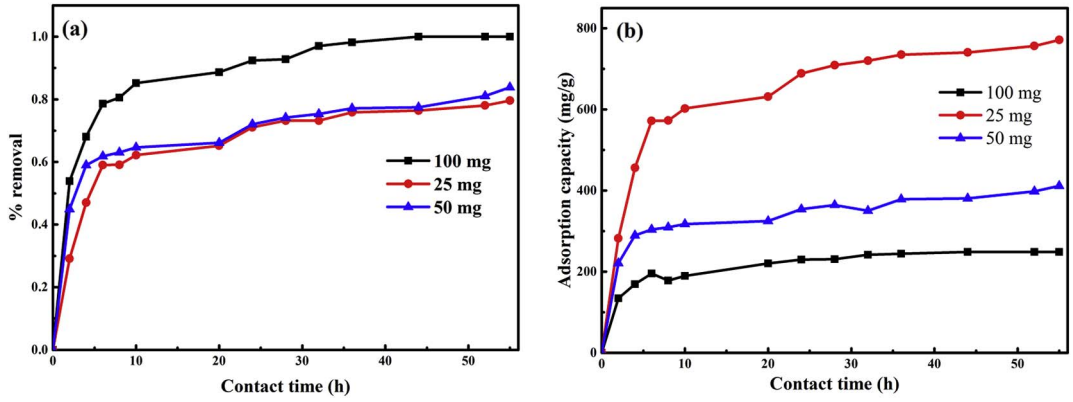

Fig. 1. The variation of (a) % removal and (b) adsorption capacity of GO versus contact time onto MgAl-MMO with different sorbent dosage. The initial solution concentration and volume is 25 mg/L and 1 L, respectively.

Download English Version:

https://daneshyari.com/en/article/8046079

Download Persian Version:

https://daneshyari.com/article/8046079

Daneshyari.com