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Removal of cadmium (II) from aqueous solution by granular activated carbon supported magnesium hydroxide

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ABSTRACT

Granular activated carbon supported magnesium hydroxide (Mg-GAC) was successfully prepared by an environmentally friendly preparation process. Effects of magnesium ion concentration on magnesium content and the removal efficiency of Mg-GAC for cadmium (II) were discussed. The raw GAC and Mg-GAC were detected by scanning electron microscopy (SEM), X-ray diffraction (XRD), and Brunauer–Emmett–Teller (BET) for comparison. The results showed that magnesium hydroxide was well distributed within the pores of GAC. From the batch experiment results, the cadmium (II) could be removed by the Mg-GAC effectively. These studies showed that cadmium (II) adsorption on GAC and Mg-GAC (magnesium content was 1.87%) was 3.47 mg/g and 8.08 mg/g, respectively with 27 mg/L initial concentration, at pH 6 and temperature 25°C, which indicated that Mg-GAC had a better adsorption capacity than untreated granular activated carbon. Besides, the adsorption data agreed well with a pseudo-second-order kinetic model.

1. Introduction

Industrial activity has notably increased the discharge of heavy metal ions into the environment, thereby resulting in undesirable pollution [1]. Cadmium is widely used in industries such as electroplating, paint pigments, plastics manufacturing, mining and metallurgical processes [2]. Cadmium is a toxic heavy metal of environmental concern and is classified as a B1 carcinogen by the U.S. Environmental Protection Agency [3]. Most of the cadmium that enters the body accumulates in the liver, kidney, pancreas and bones, which causes diseases such as anemia, hypertension, neuralgia, nephritis and secretion disorder [4]. At present there are various procedures for the removal of heavy metal ions from aqueous media such as precipitation [5], adsorption [6], ion-exchange [7], filtration [8], electrochemical technique [9], and reverse osmosis [10].

The adsorption with activated carbon is widely used to remove contaminants in water treatment processes because of its huge specific surface area and well-developed pore structures [11]. But the absorptive capacity of activated carbon is limited. Many researchers have shown that the modification of GAC could lead to increased adsorption capacity due to chemisorption of contaminants onto the surface of GAC. Activated carbon impregnated with copper salt showed an enhanced NO_2 adsorption at room temperature [12]. The use of ZnS:Cu nanoparticles loaded on activated carbon to remove Methylene blue and Auramine-O dyes from aqueous solutions showed that a small amount of the adsorbent was able to remove more than 99.5% of both dyes rapidly with high adsorption capacity in single component system and in binary-component system [13]. Iron-impregnated granular activated carbon was successfully utilized to remove arsenic [14–17]. Polyacrylamide/activated carbon hydrogel was synthesized, and its absorptive capacity was estimated using copper (II) ions and direct red 80 dye as adsorbates, which indicated that the adsorbent was efficient and useful in multi-component adsorption system [18]. An activated carbon-based multicarboxyl adsorbent has been used to remove cationic dyes from aqueous solutions, and it selectively removed the cationic dyes from aqueous solution [19].

Besides, some researchers reported that granular activated carbon supported magnesium was used to remove contaminants. GAC/MgO composite used as a catalyst effectively increased the degradation and mineralization efficiency of catechol in the ozonation process [20]. The application of GAC/MgO composite for destructive adsorption of benzene from air showed that although

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the impregnation of GAC with MgO resulted in a decrease of specific surface area and pore volume, the adsorption capacity of the GAC/MgO composite improved to around 12 times that of GAC for the removal of benzene [21]. Besides, magnesium was introduced onto the activated carbon surface via two oxygen atoms bridge and the adsorbent was used to remove Zn (II) and Cd (II) [22]. The removal efficiency of GAC/MgO composite above is efficient for contaminants.

However, some of the preparation processes above need to calcine at high temperature, which needs a huge amount of energy or is not environmentally friendly, and some need complex technology. Therefore, developing an environmentally friendly modification process with simple technology is necessary.

Magnesium hydroxide is an environmentally friendly chemical product. Due to its non-toxicity and low cost, magnesium hydroxide is an approved drug and food additive, and it has been widely used in flame-retardant composite formulations, wood pulp bleaching and cultural heritage conservation [23]. Besides, previous studies by our group on removing turbidity and reactive orange with magnesium hydroxide were made [24,25]. Magnesium hydroxide has also been widely used to adsorb heavy metals from wastewater [26]. Therefore, granular activated carbon can be modified by magnesium hydroxide to adsorb cadmium, which can use the advantages of activated carbon and magnesium hydroxide fully.

The main aim of this study was to develop an environmentally friendly preparation process of Mg-GAC with simple technology and characterize Mg-GAC to determine the amount, distribution, morphology, and species of impregnated magnesium. Finally the impacts of the amount of impregnated magnesium on cadmium adsorption capacity and efficiency and adsorption kinetic studies were investigated.

2. Experimental

2.1. Materials

The anthracite coal base of activated carbon was purchased from Ningxia Huahui Activated Carbon Company Limited with granular shape. The granular activated carbon (GAC) was selected from 1 mm to 1.6 mm in diameter.

All chemicals used in this study, including magnesium chloride, cadmium nitrate, hydrochloric acid, and sodium hydroxide, were of reagent grade. Synthetic cadmium contaminated aqueous solutions was prepared with cadmium nitrate in de-ionized (DI) water.

2.2. Preparation of adsorbents

The GAC was thoroughly washed using de-ionized (DI) water to clean impurities and powder, dried at 105 °C till the constant weight was observed, and stored in a desiccator. Regent grade magnesium chloride hexahydrate was solved in de-ionized water to prepare 0.5, 1.0, 1.5 and 2.0 mol /L solution. The magnesium solution was mixed with de-ashed GAC with the ratio of 150 mL magnesium solution per 10 g activated carbon, and the mixture was agitated at 25 °C and 180 rpm for 3 h to impregnate the magnesium (II) ions into GAC. Then, the GAC was separated from the magnesium solution and placed in an oven at 105 °C for 4 h. At the end, The GAC supported magnesium (II) ions reacted with 1 mol/L NaOH for 3 h. The Mg-GAC was washed thoroughly using DI-water and dried at 105 °C. The produced Mg-GAC was stored for further uses.

2.3. Characterization of adsorbents

The acid extraction method [27] was selected to determine the magnesium content. Three grams of Mg-GAC was added into a 250 mL conical flask containing 100 mL pH = 1 HCl solution and shaken for 10 h. Then, the Mg-GAC was separated from the solution using 0.45 μm membrane filter. The magnesium concentration in the filtrate was analyzed using the ion chromatograph (ICS-1500; DIONEX; USA). The magnesium content was calculated according to the Eq. (1).

$$\text{magnesium content} = \frac{\text{mass of magnesium}}{\text{mass of GAC} + \text{mass of magnesium}} \times 100\% \quad (1)$$

The surface distribution and morphology of the adsorbents were visualized using the scanning electron microscopy (SEM) method (Quanta 200; FEI; Czech). The X-ray diffraction (XRD) technique (vctima IV; Rigaku corporation; Japan) was carried out to determine magnesium species. The specific surface area and pore volume were determined using a nitrogen gas adsorption analyzer (ASAP 2020; Micromeritics; USA) based on the Brunauer–Emmett–Teller (BET) method.

Table 1
Magnesium content of Mg-GACs.

Mg-GACs	Mg-GAC$_1$	Mg-GAC$_2$	Mg-GAC$_3$	Mg-GAC$_4$
Magnesium ion concentration (mol/L)	0.5	1.0	1.5	2.0
Magnesium content of Mg-GAC (%)	1.31	1.87	1.52	1.08

2.4. Adsorption test

Series of experiments were carried out using 250 mL conical flasks. 300 mg Mg-GAC of different magnesium content and 100 mL water solution containing 27 mg/L cadmium ion were distributed to a series of 250 mL conical flasks. Then the conical flasks were placed in a shaker at the desired temperature for different time. After that, Mg-GAC was separated from the solution through a 0.45 μm membrane filter. Filtrate was preserved for analysis.

3. Results and discussion

3.1. Characterization of Mg-GAC

3.1.1. Magnesium content

Table 1 showed the magnesium content of Mg-GAC which was created under different conditions. As we can see, the magnesium content of Mg-GAC increased firstly and then decreased as the concentration of magnesium chloride increased.

3.1.2. SEM characterization

Fig. 1 presented the SEM images of the GAC and the Mg-GAC. As shown, both the GAC and Mg-GAC presented the inhomogeneous porous structure. Little amounts of deposited sediments could be found in the surface pores of the GAC. After modification, an increased amount of deposited sediments could be observed in the pores of GAC. And from Fig. 1a–e, the amount of deposited sediments increased firstly and then decreased, which was consistent with the results of the magnesium content determined with the acid extraction method. In conclusion, Fig. 1 clearly revealed that the magnesium uniformly deposited in the surface pores of the GAC, thereby the surface morphology of the activated carbon had been majorly modified after impregnation with magnesium.

3.1.3. XRD characterization

To evaluate the species of impregnated magnesium, Mg-GAC was analyzed by X-ray diffraction analyses and compared to GAC. Fig. 2 showed the XRD spectra of Mg-GAC and GAC. The broad diffraction peak at 43.6° of GAC was attributed to the reflections