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A novel conversion of the groundwater treatment sludge to magnetic particles for the adsorption of methylene blue

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HIGHLIGHTS

- The low-content iron sludge was converted to MPs via a simple solvothermal process.
- MPs have high efficiency of the MB removal and can be easily separated from the treated water.
- Impurities, such as Al, Si in the iron sludge, have little effect on magnetic separation of MPs.

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ABSTRACT

Iron sludge, produced from filtration and backwash of groundwater treatment plant, has long been considered as a waste for landfill. In this study, iron sludge was reused to synthesize Fe_3O_4 magnetic particles (MPs) by using a novel solvothermal process. Iron sludge contained abundant amounts of silicon, iron, and aluminum and did not exhibit magnetic properties. After treatment for 4 h, the amorphous Fe in iron sludge was transformed into magnetite Fe_3O_4, which could be easily separated from aqueous solution with a magnet. The prepared particles demonstrated the intrinsic properties of soft magnetic materials and could aggregate into a size of 1 μm. MPs treated for 10 h exhibited excellent magnetic properties and a saturation magnetization value of 9 emu/g. The obtained particles presented the optimal adsorption of methylene blue under mild conditions, and the maximum adsorption capacity was 99.4 mg/g, which was higher than that of granular active carbon. The simple solvothermal method can be used to prepare Fe_3O_4 MPs from iron sludge, and the products could be applied to treatment of dyeing wastewater.

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1. Introduction

In groundwater treatment plants, iron sludge precipitated from backwash wastewater via single-stage biofiltration contains low iron and numerous impurities of aluminum, sand, and broken filter materials. By contrast to sludge with high iron content produced in traditional two-stage aeration-filter systems, sludge with low iron content cannot be directly used to prepare red iron oxide. Aquatic pollution caused by leaching of metal ions with rain at dumping sites of iron sludge has attracted attention [1,2]; the leached ions may contaminate soil and surface water and threaten the environment [3,4]. In preventing environment pollution, iron sludge is dewatered through flocculation, coagulation, and filter pressing before disposal to landfills but this process consumes space and requires additional costs.

Iron sludge can be recycled to prepare magnetic adsorbents, but its low contents of iron and other components complicate the in situ synthesis of magnetic materials. Such materials could be prepared using high-iron-content wastes, such as red mud in alumina refineries [5,6] and electroplating sludge in plating factories [7]. Akin et al. [8] leached ferric ions from red mud through microwave digestion and used these ions to prepare nano-size Fe_3O_4 through alkaline co-precipitation. Despite the effective adsorption of arsenic ions in groundwater by the prepared Fe_3O_4-NPs, their low leaching efficiency and secondary contamination from leached acid sludge residues limit their application. High-temperature reduction is used to recover ferric irons from iron sludge. Samouhos et al. [6] used red mud with 43% iron content from an alumina refinery as a raw material to prepare a magnetic concentrate with 69.3 wt.% metallization degree; the experiment

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was performed by adjusting the microwave treatment time and the amount of brown coal in the reductive microwave process. Zhu et al. [9] also used red mud with an iron content of 50.6% (in TFe) as a raw material to synthesize magnetic particles (MPs); synthesis was conducted through reductive roasting method with anthracite as the reducing agent. The efficiency of the recycled magnetic material can reach 94.1% of the total Fe in the presence of a magnetic field. The high-temperature reductive method could be used to treat sludge with high iron content. However, this process may not be efficient for sludge with low iron content because the amount of residues increases by adding a reducing agent and brown coal or anthracite.

As an easy and controllable method, solvothermal synthesis has been widely used to prepare magnetic Fe_3O_4 particles with different sizes and morphologies in aqueous solutions [10,11]. The target product can be synthesized through solvothermal method using reductive polyol and appropriate additives in an aqueous solution [12]. Nevertheless, similar or better reductive effect of solvothermal method on iron sludge containing impurities remains undetermined. This study is the first to conduct solvothermal synthesis of Fe_3O_4 MPs from iron sludge produced in a groundwater treatment plant by using ethylene glycol as solvent and reducing agent. The adsorption of methylene blue (MB) onto the obtained MPs was then investigated.

2. Materials and methods

2.1. Materials

Backwash wastewater was collected from a groundwater biofilter of Jilin YaTai Cement Company (China) and then precipitated for 8 h. After precipitation, the settled yellowish brown iron sludge was collected and dried at 80 °C for 2 h in a vacuum-drying oven.

Anhydrous sodium acetate, ethylene glycol, and MB were purchased from Sinopharm Chemical Reagent Co., Ltd. (Beijing, China). Powder active carbon (PAC) and granular active carbon (GAC) were supplied by Tianjin Fuchen Chemical Reagents Factory (Tianjing, China). All chemicals and reagents were of analytical grade.

2.2. Preparation process

MPs were prepared using a one-step solvothermal method. About 0.675 g of dried iron sludge was mixed with 20 mL of ethylene glycol (used as solvent and reducing agent) under magnetic stirring and then added with 3.6 g of sodium acetate as a ligand. After stirring for 30 min, the suspension became yellowish brown and was transferred into a teflon-lined stainless-steel reaction kettle. The kettle was sealed and calcined at 180 °C for 2 h–20 h with a heating rate of 4 °C/min in a drying oven. After the solvothermal process, the autoclave was cooled to room temperature and black Fe_3O_4 particles at the bottom of the teflon were collected. The particles were ultrasonically washed five times with deionized water, collected with a magnet after each washing, and vacuum dried at 40 °C overnight. The effect of solvothermal treatment time on iron sludge was investigated by varying the time from 2 h to 20 h, and the obtained particles were denoted as "MP-*x*" (*x* represents the solvothermal treatment time).

2.3. Adsorption measurement

MB was used as the substrate to evaluate the adsorption capacity of the obtained MPs. About 20 mL of MB (10, 30, 60, 100, 150, 200, and 300 mg/L) was mixed with 14 mg of MPs in an Erlenmeyer flask. The flask was sealed with parafilm and shaken at 200 rpm and 25 °C in an incubator. The initial pH of the mixture was 6.8 and was not adjusted during the experiment. After 2 h, the flask was removed from the incubator and MPs were separated from the solution by using an external magnetic field. The supernatant was filtered with a 0.45 μm filter, and the filtrate was collected to determine MB concentrations. PAC and GAC were used as controls. All treatments were performed in triplicates, and data were averaged.

2.4. Material characterization

Magnetization was measured at room temperature by using a magnetometer (Quantum Design, USA) with a SQUID-VSM system. X-ray diffraction patterns were determined with a diffractometer (Rigaku, Japan) using Cu Kα radiation and 2θ range of 10°–70°. The composition of iron sludge was determined using X-ray fluorescence (Rigaku, Japan), and cations in MPs were investigated through microwave digestion according to the EPA method 3051 A [13]. Transmission Mössbauer spectroscopy experiments were carried out using an MP500 spectrometer (Oxford, Britain) at room temperature with α-Fe^0 as a reference. The valence states of Fe and O on the particle surface were determined through X-ray photoelectron spectroscopy (VG-ADES, Britain) with an Mg Kα X-ray source at a residual gas pressure lower than 10^{-8} Pa. The surface characteristics of MPs were observed with a field-emission scanning electron microscope (FE-SEM; FEI Co., USA) with an accelerating voltage of 200 kV. The specific surface areas of MPs were determined through nitrogen adsorption–desorption measurements (TriStar 3000).

The concentration of MB in the filtrate was determined using a UV–vis spectrophotometer (Purkinje General, China) with OD at 655 nm. The amount of adsorbed MB (q_e (mg/g)) was calculated using Eq. (1):

$$q_e = (C_0 - C_e)V/m \quad (1)$$

where C_0 and C_e represent the initial and equilibrium concentrations of MB (mg/L), respectively, V denotes the total volume of MB solution used for analysis (L), and m is the mass of MPs (g).

3. Results and discussion

3.1. Magnetic measurement

The magnetic hysteresis loops determined at room temperature for iron sludge and MPs are shown in Fig. 1(c). The iron sludge precipitated from backwash wastewater through single-stage filter did not exhibit magnetism as indicated by its inability to move in the magnetic field. After solvothermal treatment for 2 h, MP-2 showed a magnetization curve similar to that of iron sludge, indicating a

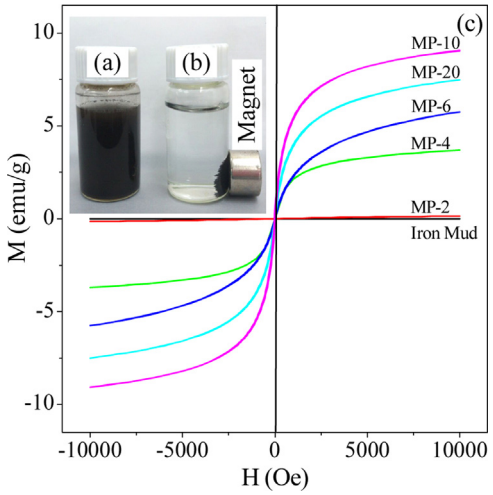

Fig. 1. (a) MP aqueous solution; (b) magnet-separated MPs; and (c) hysteresis loops of iron sludge and MPs.

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