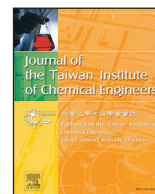




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Efficient removal/recovery of Pb onto environmentally friendly fabricated copper ferrite nanoparticles

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ABSTRACT

Copper ferrite (CuFe_2O_4), a novel type of nanoparticle manufactured from industrial sludge, has been found to be effective for Pb remediation in aqueous solutions. Our results indicated that over 90% of Pb was removed/recovered from solutions at pH 4.5 and 298 K. A significant negative relationship was detected between the amount of Pb adsorbed and the concentrations of NaCl, indicating that the Pb adsorption onto CuFe_2O_4 was dominated by the outer-sphere complex. This relationship successfully showed a fast kinetics process during Pb adsorption with the implementation of kinetic models, while the simultaneous monolayer and multilayer mechanisms in Pb adsorption could be well described using isotherm models. Additionally, in simulated seawater adsorption, Pb adsorption was independent of coexisting cations, thereby confirming the Pb removal/recovery potential of CuFe_2O_4 in the seawater matrix. In desorption experiments, the recovering sequence of desorption reagents was $\text{HNO}_3 \cong \text{HCl} > \text{H}_2\text{SO}_4 > \text{H}_3\text{PO}_4$. This result indicated that Pb desorption efficiency was highly dependent upon reagent concentration, and 0.005 N HNO_3 was sufficient for Pb desorption. CuFe_2O_4 is a good and low-cost solution for simultaneously resolving industrial sludge and remediating aqueous Pb.

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1. Introduction

Lead (Pb) is regarded as one of the most toxic substances because it can damage the kidney, liver, reproductive, mental, and central nervous systems [1–5]. Pb is mainly discharged into the environment through effluents of the steel, battery, textile, fertilizer manufacturing, and automotive industries [6,7]. The growing usage and production of Pb implies a hazardous potential for its increased release and distribution in the natural environments. The maximum levels for Pb in drinking water are 15.0 and 10.0 $\mu\text{g/L}$ according to the US Environmental Protection Agency (US EPA) and World Health Organization (WHO), respectively [8]. However, much higher concentrations (hundreds of $\mu\text{g/L}$ to tens of mg/L) can be found in aquatic systems because of anthropogenic activities. If there is no suitable method for treating Pb anthropogenic contamination, human health and aquatic ecosystems will be threatened. Because of the problems mentioned above, research interest has

intensified in the development of proper technology for Pb removal from solutions.

Approaches to remove Pb from waters, such as chemical precipitation, ion exchange, electrochemical techniques, oxidation, and adsorption, have been widely investigated. Nevertheless, most available technologies may have technical and/or economical defects (i.e., sensitivity to operational conditions, sludge generation, and/or high operational costs). Adsorption is recognized as an effective, simple, and economic method for the removal of heavy metals from solutions [9–16]. In addition, the reversibility of the adsorptive reaction allows an adsorbent to recycle and regenerate as well as improves environmental protection. Many studies have focused on the removal of Pb using functional modified or composite adsorbents [17–19]. For example, Zhang et al. [17] studied the removal of Pb from aqueous systems using the relatively high loading of the amino-functionality of $\text{Fe}_3\text{O}_4@ \text{SiO}_2\text{-NH}_2$. The result demonstrated that the maximum Pb adsorption occurred in the range of pH 5–6 with a maximum adsorption capacity of 238 mg/g at 298 K [17]. Another amino-functionalized magnetic nano-adsorbent (MNPs- NH_2) investigated by Tan et al. [18] showed that the adsorption of Pb was found to follow the Langmuir isotherm, and the maximum adsorption capacity was 40.10 mg/g at pH 5 and 298 K. Zhang et al. [19] reported that the fried egg jellyfish-like $\gamma\text{-AlOOH(Boehmite)}@ \text{SiO}_2/\text{Fe}_3\text{O}_4$ porous magnetic

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microspheres can effectively remove Pb, and the maximum adsorption capacity, $q_m = 214.59$ mg/g, is approximately 11.7-fold and 34.6-fold higher than $\text{SiO}_2/\text{Fe}_3\text{O}_4$ and Fe_3O_4 magnetic microspheres, respectively. Although these adsorbents have a high adsorption capacity of Pb, the complex manufacturing processes and the high cost of these adsorbents may impede their industrial development.

Copper ferrite nanoparticles (CuFe_2O_4) with a spinel structure have a cubic close-packed arrangement of oxygen ions with Cu^{2+} and Fe^{3+} ions at two different crystallographic sites [20,21]. CuFe_2O_4 has been reported to remove contaminants such as acid orange II [22], Cd [23], and As [24] from different solutions. However, the cost of CuFe_2O_4 manufactured using the sol-gel method, auto-combustion, or co-precipitation [25–27] is still high when the raw materials are adopted from pure chemicals. If the raw materials can be obtained from industrial sludge, then the cost can be drastically reduced.

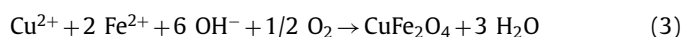
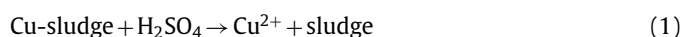
This research applies synthesized CuFe_2O_4 extracted from the sludge of printed circuit boards (PCBs) to assess the remediation of Pb from solutions and the performance of CuFe_2O_4 . Our previous study successfully recycled copper powder from PCB sludge using a combination of acid leaching and chemical exchange [28]. After the combination of these technologies, the ferrite process was conducted to ensure that both the supernatant and the sludge can meet environmental regulations. Thus, the sludge generated from the ferrite process is regarded as a novel adsorbent and will be used for testing its capability and efficiency of Pb removal.

This series of batch experiments will study the following: (i) the mechanistic, kinetics and equilibrium exchange of Pb during the sorption processes; and (ii) the removal/recovery of Pb using the synthesized CuFe_2O_4 nanoparticles. Additionally, the physical and chemical properties, such as adsorbent crystalline phase, saturation magnetization, point of zero charge, and specific surface area, were carefully examined. The effect of solution pH, ionic strength and coexisting cations on Pb adsorption kinetics and isotherms were also calculated and discussed. The information obtained implies the potential development of an effective adsorbent for the uptake of Pb using copper ferrite (CuFe_2O_4) nanoparticles.

2. Materials and methods

2.1. Manufacture of copper ferrite (CuFe_2O_4) nanoparticles

The details for the preparation of copper ferrite (CuFe_2O_4) nanoparticles are given in our previous study [28]. The procedures were conducted in the following order: acid leaching of the PCB sludge, chemical exchange of Fe and Cu, and the ferrite process. Approximately 10 L of 2 N sulfuric acid was added to 500 g of PCB sludge for Cu extraction at a temperature of 323 K for 60 min. Fe^0 was used as the sacrificial metal to substitute Cu^{2+} in the liquids during the chemical exchange reaction. To ensure that the supernatant fulfills the effluent standards, the ferrite process was performed after the chemical exchange. The novel, low-cost adsorbent CuFe_2O_4 nanoparticles were thus manufactured after the ferrite process. The reactions of acid leaching, chemical exchange, and the ferrite process can be expressed as Eqs. (1)–(3), respectively:



The adsorbent was collected using a magnetic separation method. The adsorbent was then washed many times with Milli-Q water until the pH of the solution reached approximately 7. The

solids were dried at 323 K for 24 h in an oven and stored for further investigation.

2.2. Characterization of the copper ferrite nanoparticles

The characteristics of the copper ferrite nanoparticles were evaluated prior to the sorption batch experiments. The Brunauer, Emmett and Teller (BET) theory was used for determining the surface area via ASAP 2010 analyzer (Micromeritics, USA) with N_2 adsorption at 77 K. The mineralogy was characterized using an X-ray diffractometer (XRD, D8 Advance, Bruker, Germany) with a graphite monochromatic copper radiation over a 2θ range of 10–80°, and then the crystalline phases could be determined using the corresponding spectrum of the standard. Scanning electron microscopy (SEM, JSM-6330, Japan) was used to examine the surface morphology of the CuFe_2O_4 particles. The saturation magnetization of the recycled copper ferrite was measured using a Superconducting Quantum Interference Device (MPMS-XL7, Quantum Design, USA) at 298 K. The point of zero charge (PZC) is the pH at which the particles in suspension have a net charge of zero and no mobility in the electric field. The determination of the PZC in this study was performed using a zeta potential analyzer (Zetasizer 2000, USA).

2.3. Chemicals

All chemicals were of analytical grade and were used without purification. Pb solution used for the batch sorption experiments was diluted from a high purity Pb solution (1000 ± 2 mg/L in $< 5\%$ HNO_3 ; manufactured by Merck, Germany). Nitric acid (97%) and sodium hydroxide, used to adjust the pH, were obtained from Sigma-Aldrich. The reagents used in the experiments were all analytical grade and Milli-Q water (18.2 M Ω /cm) was used for dilution. The bottles used in this study were all acid washed before each experiment.

2.4. Batch adsorption procedure

To evaluate the equilibrium states of Pb during the adsorption process, a series of batch experiments were conducted. The detailed experimental procedures are described as follows. 10 mL of Pb solution and a fixed amount of CuFe_2O_4 nanoparticles were added to 15-mL centrifuge tubes. The centrifuge tubes were then placed on the shaft of a rotary shaker after tightening the caps. The rotation speed was set at 30 rpm, and the temperature was controlled at 298 ± 1 K. The metal uptake q_t (mg/g) was determined using

$$q_t = \frac{C_0 - C_t}{m_{\text{ads}}} \quad (4)$$

where C_0 and C_t are the metal concentration in the liquid phase at the initial and at time t (mg/L), respectively; m_{ads} is the adsorbent amount remaining in the solution (g/L).

To avoid the precipitation of Pb, pH was controlled at 2.50–4.50 by adding 0.1 N NaOH or 0.1 N HNO_3 solution. In the adsorption kinetics and isotherms study, the reaction equilibrium was monitored using a time scale ranging from 3–2280 min, and the adsorption behavior during the reactions was investigated using initial Pb concentrations of 20–700 mg/L. Furthermore, the NaCl range of 0.001–1.0 N was considered in regard to the influence of ionic strength because the solution matrix will affect the Pb adsorption efficiency. In addition, the simulated sea salts dissolved in Milli-Q water were used to determine whether Pb adsorption would be affected by other cations. All experiments provided in this study were performed in triplicate. The relative standard deviation (RSD) of three replicate analyses was lower than 3%. The solid and liquid

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