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ORIGINAL ARTICLE

Removal of copper, nickel and zinc by sodium dodecyl sulphate coated magnetite nanoparticles from water and wastewater samples

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Abstract In the present study, sodium dodecyl sulphate-coated Fe_3O_4 nanoparticles (SDS–Fe_3O_4 NPs) were applied to remove Cu(II), Ni(II) and Zn(II) ions from water and wastewater samples. The effects of pH of solution, SDS, Fe_3O_4 NPs and salt addition on removal efficiency of the metal ions were investigated and optimized. Salt addition has a negative effect on the removal efficiency of the metal ions, thus extraction follows the ion exchange mechanism. The results showed that the adsorption process onto the adsorbent is very fast under optimum conditions and nearly 1 min of contact time was found to be sufficient for completion of the metal ions' adsorption. Adsorption equilibrium of the metal ions reveals that data were fitted well to the Langmuir isotherm. Also, the maximum monolayer capacity, q_{max}, obtained from the Langmuir isotherm was 24.3, 41.2 and 59.2 mg g^{-1} for Cu(II), Ni(II) and Zn(II), respectively. Desorption experiments by elution of the adsorbent with methanol show that the SDS-Fe_3O_4 NPs could be reconditioned without significant loss of its initial properties even after three adsorption–desorption cycles. Finally, application of the SDS-Fe_3O_4 NPs as efficient adsorbent material for removal of the metal ions from Iran Khodro's wastewater samples was investigated and satisfactory results were obtained.

1. Introduction

Heavy metals released into the environment pose a significant threat to the ecosystem and public health because of their toxicity and persistence (Seiler et al., 1998). Therefore, heavy-metal contamination is still an important problem in both developing and developed countries throughout the world (Danazum and Bichi, 2010; Inglezakis et al., 2003; Momodu and Anyakora, 2010).

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Various methods have been developed for the removal of heavy-metal ions from wastewater which include chemical precipitation/coagulation, membrane technology, electrolytic reduction, ion exchange and adsorption. The advantages and disadvantages associated with each method are reviewed elsewhere (O'Connell et al., 2008). The adsorption process is one of the most common methods used for the removal of heavy-metal ions because of its simplicity, convenience, and high removal efficiency (Stafiej and Pyrzynska, 2007; Afkhami et al., 2007; Santos Yabe and de Oliveira, 2003; Cervera et al., 2003). The adsorption on activated carbon is attractive to many scientists due to the effectiveness of the removal of heavy-metal ions in trace quantities (Rao et al., 2009). However, the process has not been used extensively because of its high cost (Rao et al., 2009; Dias et al., 2007; El-Shafey et al., 2002). Therefore, the use of alternative low-cost materials as potential adsorbents for removal of heavy metals has been emphasized recently (Gupta et al., 2009). On the other hand, the development of heavy-metal adsorbents with high adsorption capacity, fast adsorption–desorption kinetics, and easy separation and regeneration is in great demand.

Recently, application of nanoparticles (NPs) for the removal of pollutants has come up as an interesting area of research (Faraji et al., 2010a). Compared to conventional adsorbents with micrometre size, nanomaterials offer a significantly higher surface area-to-volume ratio and a short diffusion route, resulting in high adsorbent capacity, rapid removal dynamics and high removal efficiencies (Klabunde, 2001). Moreover, magnetic nanoparticles (MNPs) can be easily separated under external magnetic fields (Faraji et al., 2010b–d; Ma'mani et al., 2010) and therefore, no sample centrifugation or filtration is needed after treatment.

In this study, MNPs have been synthesized using a co-precipitation method and after modification of them with sodium dodecyl sulphate (SDS), they were applied for the removal and recovery of copper, nickel and zinc from industrial wastewater due to high concentration of these metal ions in the real samples. The objectives of this study are: (1) to assess the performances of MNPs for the removal of the heavy-metal ions, (2) to achieve the possible regeneration of MNPs for reuse, and (3) to explore the mechanisms of metal adsorption onto MNPs.

2. Experimental section

2.1. Chemicals and reagents

All reagents were of analytical reagent grade and were used as supplied. Stock solutions (1000 mg L^{-1}) of Cu(II), Ni(II) and Zn(II) were prepared by direct dissolution of proper amounts of $Cu(NO_3)_2{\cdot}3H_2O$, $Ni(NO_3)_2{\cdot}2H_2O$ and $Zn(CH_3COO)_2{\cdot}2H_2O$ salts from Merck (Darmstadt, Germany) in doubly distilled water. The standard solutions were diluted with doubly distilled water to prepare the mixed standard solutions. Ferric chloride ($FeCl_3{\cdot}6H_2O$), ferrous chloride ($FeCl_2{\cdot}4H_2O$), sodium hydroxide, sodium dodecyl sulphate (SDS), methanol, ethanol, sodium chloride and hydrochloric acid were obtained from Merck. The pH of the solutions was adjusted by dropwise addition of nitric acid (0.01 mol L^{-1}) and/or sodium hydroxide solutions (0.01 mol L^{-1}).

2.2. Apparatus

Concentrations of the heavy metals in all samples were determined using a Varian SpectrAA 200 (Springvale, Victoria, Australia) atomic absorption spectrometer (AAS). Hallow cathode lamps at respective wavelengths (resonance line) were used as radiation sources for each metal ion. An air–acetylene flame was used for determination of the metals ions. All instrumental settings were performed based on the procedure recommended by the manufacturer. Magnetic separation was fulfilled by a supermagnet with 1.2 Tesla magnetic field, N 35 model (5 × 3 × 2 cm) from Tehran Magnet (Tehran, Iran). A Heidolph mechanical-stirrer RZR 2102 control model (Kelheim, Germany) was applied for stirring of the metal ion solutions with a glassware stirrer. The pH values of solutions were determined and adjusted using a pH meter model WTW (Inolab, Germany) with a combined glass-calomel electrode.

2.3. Synthesis of Fe_3O_4 NPs

Fe_3O_4 NPs were prepared by the chemical co-precipitation method *via* a reactor designed in our previous work (Faraji et al., 2010b). Briefly, 10.4 g of $FeCl_3{\cdot}6H_2O$, 4.0 g of $FeCl_2{\cdot}4H_2O$ and 1.7 mL of HCl (12 mol L^{-1}) were dissolved in 50 mL of deionized water in order to prepare the stock solution of ferrous and ferric chloride in a beaker which was then degassed by nitrogen for 20 min. Simultaneously, 500 mL of 1.5 mol L^{-1} NaOH solution was degassed (for 15 min) and heated to 80 °C in a reactor. Then, the stock solution was added dropwise using the dropping funnel during 30 min under nitrogen gas protection and vigorous stirring (1000 rpm) by a glassware stirrer. During the whole process, the solution temperature was maintained at 80 °C and nitrogen gas was purged to remove the dissolved oxygen. After the completion of reaction, the obtained Fe_3O_4 NPs precipitate was separated from the reaction medium by magnetic field, and then washed with 500 mL doubly distilled water four times. Finally, the obtained Fe_3O_4 NPs were re-suspended in 500 mL of degassed deionized water. The synthesized Fe_3O_4 NPs in this work as same as our previous work (Faraji et al., 2010b), were previously characterized by using IR, SEM, and XRD techniques (Faraji et al., 2010c).

2.4. Optimization of metal ion adsorption

Optimization studies were carried out according to the following procedure: (1) 20 mL aqueous solution of the metal ion (10 mg L^{-1}) was prepared in a 50 mL glass beaker by addition of the appropriate amount of the metal ion stock solutions; (2) 1 mL of the Fe_3O_4 NPs suspension (containing 10 mg of Fe_3O_4 NPs) was added to the metal ion solution; (3) pH of the solution was adjusted to the desired value and then SDS was added into the metal ion solution; (4) the mixed solution was stirred for a desired time; (5) after metal ion adsorption, Fe_3O_4 NPs were magnetically separated from the sample solution; (6) the residual metal ion concentration in the supernatant clear solution was determined by AAS using a calibration curve. The following equation was applied to calculate the metal ion removal efficiency in the treatment experiments:

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