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Synthesis and characterization of core-shell magnetic nanoparticles $Fe_3O_4@SiO_2$ functionalized with theophylline molecules as an effective and recyclable nanoadsorbent for the removal of Pb^{2+} ions from aqueous solutions

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1. ABSTRACT

In this study, $Fe_3O_4@SiO_2$ core-shell magnetic nanoparticles were synthesized using coprecipitation and Stöber methods. Afterwards, the obtained nanoparticle is functionalized with organic molecules and finally theophylline. The synthesized nanoparticle was used as an effective adsorbent for the removal of lead ions from aqueous solutions. Then, the structural characteristics, size and morphology of the synthetic nanoadsorbent were determined using analyses such as Fourier transform infrared spectroscopy, X-ray diffraction, transmission electron microscopy, scanning electron microscopy, particle size distribution, nitrogen gas adsorption-desorption, thermogravimetric analysis, X-ray energy dispersive spectroscopy and vibrating sample magnetometer. The effective parameters in the nanoadsorbent adsorption process are evaluated by investigating of contact time, pH, initial concentration of Pb^{2+} ions and adsorbent dosage. The results indicate that the maximum Pb^{2+} adsorption capacity of 130.5 mg/g occurs when 25 mg of $Fe_3O_4@SiO_2$-TCT-Theophylline nanoadsorbent, 50 mL of Pb^{2+} solution, initial concentration mmol/L0.35, at pH=7 and a contact time of 24 min. In addition, the synthetic nanoadsorbent has the ability to be recycled and reused in sequential adsorption-desorption processes for 6 times without serious reduction in adsorption activity.

Keywords: $Fe_3O_4@SiO_2$ Nanoparticles, Theophylline, Magnetic Adsorbent, Effective Adsorption, Lead Ion, Sequential Adsorption-Desorption.

2. INTRODUCTION

Heavy metal ions are considered as a serious threat to living organisms and human health due to their high stability and bioaccumulation in food. These metals cause numerous problems even at low concentrations. Therefore, their removal from industrial wastewater and effluents such as petrochemicals, refineries, fertilizer factories and pulp and paper mills has attracted the attention of researchers and the global community due to their significant toxic effects on the surrounding environment. Lead is one of the toxic and dangerous metals. Its entry into the human body cause complications such as cancer, kidney and heart diseases, anemia, memory loss, hallucinations, and possible damage to DNA [1, 2].

In recent years, various methods such as ion exchange, extraction, membrane filters, chemical precipitation, adsorption and electrochemical methods have been used to separate heavy metal ions from aqueous solutions. Moreover, various methods such as reverse osmosis, evaporation, adsorption, ion exchange, coagulation, electrochemical and sedimentation have been applied to recover heavy metals from wastewater and industrial effluents. Among the methods for separating

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heavy metal ions, the use of adsorption methods has received much attention due to its significant features and advantages such as low energy consumption, low cost, high adsorption capacity, high efficiency, easy operation and simple maintenance. Functional groups play a fundamental role in the functional ability of the adsorbent by forming chemical complexes with metal ions [3, 4].

3. MATERIALS AND METHODS

The chemicals and compounds used for the synthesis of nanosorbents were purchased from Aldrich. In order to investigate the structure of the synthetic nanoparticles, X-ray diffraction (Bruker AXS D8-advance X-ray diffractometer) with Cu Kα radiation and a wavelength of 1.5418 nm was used. The morphology of the synthetic samples was examined using a scanning electron microscope (FE-SEM, HITACHI S-4160). The particle size distribution in the synthetic samples was evaluated using a HORIBA-LB550 device. The particle size in the synthetic samples was examined using a Philips EM208 device (100 kW voltage boost). Fourier transform infrared spectroscopy (FT-IR, Shimadzu FT-IR 8300) was used to investigate the steps of the nanoparticle synthesis. The thermal stability of the synthetic nanoparticles was investigated using a thermal analysis device (TGA, Perkin Elmer instrument) at a rate of 20 ^{o}C/min and under nitrogen gas. The magnetization properties of the samples were investigated using a vibrating sample magnetometer (VSM, Meghnatis Daghigh Kavir Co., Iran). Energy dispersive X-ray (EDX, Philips scanning electron microscopy) was used to investigate the type of elements in the synthetic nanoadsorbent. The specific surface area and porosity of the synthetic samples were investigated using the Brunauer-Emmett-Teller (BET) method and nitrogen adsorption-desorption isotherm. The concentration of lead ions in the solution was determined using inductively coupled plasma (ICP).

In the present study, core-shell Fe_3O_4@SiO_2 nanoparticles were first synthesized using coprecipitation and Stöber methods. Afterwards, functionalization of Fe_3O_4@SiO_2 nanoparticles with 3-aminopropyltriethoxysilane, trichlorotriazine, 3-bromopropylamine and theophylline molecules was performed. The proposed nanostructure was investigated as an effective adsorbent for the adsorption of lead ions after characterization using FT-IR, XRD, TEM, FE-SEM, DLS, BET, EDX, EDX, TGA and VSM techniques [5].

4. RESULTS AND DISCUSSION

In order to investigate the performance of the presented adsorbent in solid phase extraction of lead ions, parameters such as adsorbent dose, pH, and initial concentration and contact time were optimized and investigated. Nanosorbent dose optimization was performed using 50 mL of lead ion solution (initial concentration of 0.35 mmol/L) at pH 7, ambient temperature, and contact time of 30 min and in a range of 5-27.5 mg of Fe_3O_4@SiO_2-TCT-Theophyline nanosorbent. According to the results, the adsorption rate of lead ions from the solution increases with increasing adsorbent amount and a maximum adsorption of 90% occurs in the presence of 25 mg of adsorbent.

The pH optimization of the solution was carried out in the presence of 25 mg of Fe_3O_4@SiO_2-TCT-Theophyline nanoadsorbent in 50 ml of solution (initial concentration 0.35 mmol/L) at ambient temperature and a contact time of 30 minutes. At low pH, due to the protonation of surface-active groups and inactivation of heteroatomic groups of the adsorbent, the adsorption rate of lead ions decreases. The adsorption rate of metal ions by the adsorbent increases with increasing pH and availability of active sites and the maximum adsorption capacity occurs at pH 7.

In order to investigate the effect of the initial concentration of lead on the adsorption rate of Fe_3O_4@SiO_2-TCT-Theophyline nanoadsorbent, experiments were carried out in the presence of 25 mg of nanoadsorbent, a concentration range of 0.4-0.1 of lead ion solution in 50 ml of solution at pH 7 and a contact time of 30 minutes. The results indicate that the maximum adsorption capacity of the nanosorbent is observed in the presence of lead ion solution with an initial concentration of 0.35 mmol/L. Increasing the initial concentration led to better performance in the adsorption of lead ions by the adsorbent which is due to the increase in the mass gradient between the lead ion solution and the nanoadsorbent.

In order to optimize the contact time of the nanosorbent in the adsorption process, 50 ml of lead ion solution with an initial concentration of 0.35 mmol/L and 25 mg of Fe_3O_4@SiO_2-TCT-Theophyline were used in the time range 4-28 minutes, ambient temperature and pH 7. According to the results, the adsorption of lead ions from the solution increases with increasing contact time. The maximum adsorption of 90% occurs after 24 minutes of the adsorption process

Please conclude your work incorporating your most important finding as well as future works (3-4 lines). The information conveyed by the extended abstract SHOULD NOT BE different from that conveyed by the original paper (in Persian).

5. CONCLUSION

In the present study, Fe_3O_4 nanoparticles were first synthesized using the co-precipitation method with a particle size of 10 nm. Afterwards, the surface coating of these nanoparticles with silica nanoparticles was performed using the Stöber method, which resulted in the synthesis of core-shell Fe_3O_4@SiO_2 nanoparticles with a particle size of about 20 nm. After functionalizing these nanoparticles with organic compounds and theophylline molecules, the desired nanosorbent with spherical morphology and particle size of 30 nm was synthesized. In the next step, the structural characteristics, particle size and morphology of this nanosorbent were determined using FT-IR, XRD, TEM, FE-SEM, DLS, BET, TGA, EDX and VSM techniques. Moreover, optimization of the effective parameters in the adsorption performance such as adsorbent dose, solution pH, and initial concentration of target ion and contact time of adsorbent with target ions was carried out. The results indicated the maximum adsorption capacity (90%) in the presence of 25 mg of Fe3O4@SiO2-TCT-Theophyline nanosorbent, 50 ml of lead solution (initial concentration of 0.35 mmol/L) at pH 7 for a contact time of 24 minutes at ambient temperature. In addition, the synthetic nanosorbent has obvious features and advantages such as easy synthesis, high adsorption rate, excellent adsorption capacity, use of small amounts, magnetic field separation capability,

ability to recycle and reuse in successive adsorption-desorption cycles. Therefore, Fe_3O_4@SiO_2-TCT-Theophyline nanosorbent has the ability to be used as a powerful and effective adsorbent for removing heavy metal ions from industrial effluents and wastewaters.

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