

Alginate-based nanocomposites for efficient removal of heavy metal ions



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ABSTRACT

Cobalt ferrite nanoparticles (CF), titanate nanotubes (T), alginate (G) and their nanocomposite microparticles (CF/G and T/G) were prepared and used for efficient removal of Cu²⁺, Fe³⁺ and As³⁺ ions from water. The nanocomposites were characterized using field emission scanning electron microscopy (FESEM), X-ray diffraction (XRD), high resolution transmission electron microscopy (HRTEM), FTIR and vibrating sample magnetometer (VSM). In addition, the effects of pH, contact time, adsorbent weight and heavy metal ion concentration on the removal efficiency were investigated. Our results revealed a successful preparation of the nanocomposite particles. The optimized batch experiment conditions were found to be pH of 6.5, contact time of 2 h and adsorbent weight of 0.15 g. The removal efficiencies for Cu²⁺ using G, CF, T, CF/G and T/G were found to be 91%, 100%, 99.9%, 95% and 98%, respectively. While that of Fe³⁺ removal was 60%, 100%, 100%, 60% and 82%, respectively. Efficient removal of As³⁺ ions was also attained (98% upon using T nanoadsorbents). The current study demonstrated that the developed nanomaterials (CF and T) and their corresponding alginate-based nanocomposite microparticles could be further tailored and used as efficient adsorbents for the uptake of different heavy metal ions from wastewater.

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1. Introduction

Contamination of water with heavy metals is one of the greatest dilemma faced the global community due to their risky effect on human health even at low concentrations [1]. The toxic heavy metals in the environment have originated from electronics, food, cosmetics, paints, plastics, herbicides, pesticides, vehicle traffic emissions, etc. [2–4]. These toxic metal ions include copper (Cu²⁺), iron (Fe³⁺) and arsenic (As³⁺) which have highly dangerous effects on the human body. The American Environmental Protection Agency (EPA) adopted that the maximum allowable limit of Cu²⁺ and As³⁺ in drinking water is 1.3 and 0.01 mg/l, respectively [5,6], while the acceptable limit of iron as recommended by WHO ranged between 0.3–3 mg/l [7].

Interestingly, the removal of these toxic metals from wastewater is an urgent requirement for providing a good health. Up to now, many common methodologies such as adsorption, precipitation, membrane filtration and ion exchange have been widely employed for heavy metal uptake from wastewater [8–10]. The removal of

heavy metals by the adsorption technique appeared to be an efficient, economical, simple and used for numerous years [11–14]. Various adsorbents such as magnetic ferrite nanoparticles, nanotitanates, alginate biopolymer and their composites were used to remove the toxic metal ions [15–24].

It is worth mentioning that applying nanomaterials in the adsorption and environmental applications have received a great consideration due to their high surface area and greater adsorption capacity [25]. Titanate nanotubes, with their high surface area, have an extraordinary adsorption capacity compared to traditional powder titanate, which have low adsorption capacity because of low surface active sites. However, the application of materials at the nanoscale for such environmental purposes is still limited due to the difficulty of separation and recovery from the treated solutions. Certain nanomaterials such as magnetic nanoparticles can be separated easily from solution with the aid of an external magnetic field [26].

Likewise, the difficulty of nanoadsorbents separation can be evaded via their incorporation into polymeric matrix, such as alginate, in the form of nanocomposites. Bio-based polymers like cellulose and chitosan have great advantages which include non-toxicity, availability, biocompatibility, biodegradability and low cost. Moreover, the hydrophilicity of these polymers introduces

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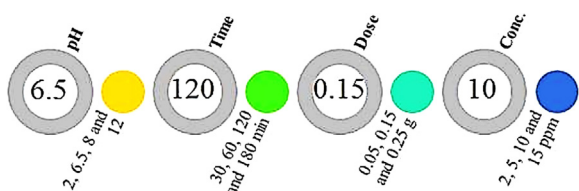


Fig. 1. Optimized conditions for metal uptake study.

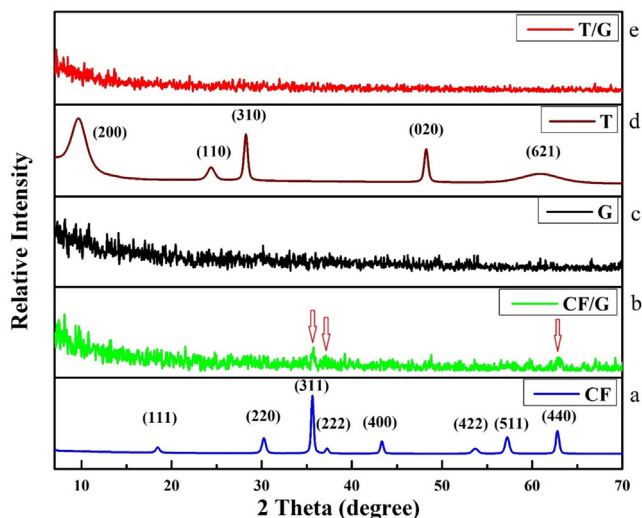


Fig. 2. XRD patterns of G, T/G, CF/G, and CF.

water fluxes higher than most of the synthetic polymers. So, they were used widely in the water purification purposes [27]. Alginate (G) is a non-toxic, biocompatible and biodegradable biopolymer which is extracted from brown seaweeds. It consists of blocks of 1–4 linked α -L-guluronic and β -D-mannuronic acids [28]. In the presence of divalent cations, especially Ca^{2+} ion, G can easily form crosslinked gel matrices. Therefore, these Ca-crosslinked G matrices can be used to prepare adsorbents in gel phase, which are easier to handle than the powder materials [13]. The use of G-based formulations as efficient adsorbents is related particularly to the presence of carboxylic groups in the G structure which enable it to form complexes with metal ions in aqueous solutions [29,30].

In the current study, cobalt ferrite nanoparticles (CF) and titanate nanotubes (T) were synthesized, and used to develop new series of alginate (G)-based nanocomposite microparticles (CF/G and T/G). The developed nanomaterials and their nanocomposite microparticles were investigated as potential adsorbents for efficient removal of Cu^{2+} , Fe^{3+} and As^{3+} ions from water. It is worth mentioning that both CF and T were synthesized and selected as the nanofillers upon preparing the G-based nanocomposites not only because of the lack of previous studies reporting their use in removal of metal ions, particularly Fe^{3+} , but also because CF and T were found, in the present study, to have tunable surface charges at different pH values which enables the improvement of adsorption efficiency towards metal ions.

2. Experimental

2.1. Materials

All the reagents were of analytical grade and used as received without further purification. $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ and NaOH were purchased from Dop Organik Kimya (Turkey), Loba Chemie laboratory reagents (India) and PioChem laboratory

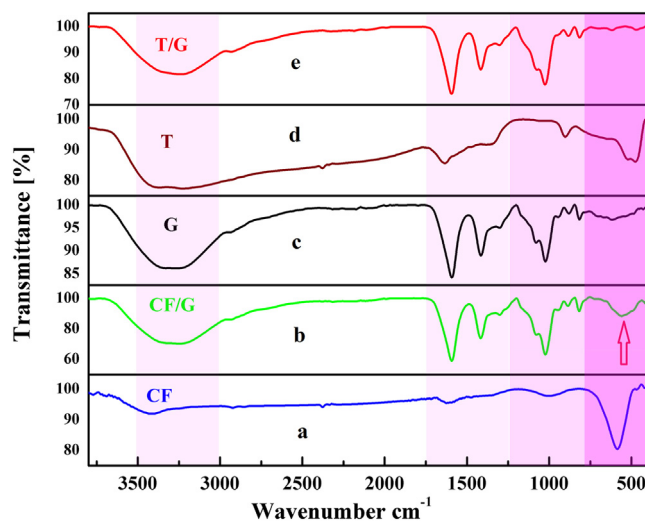


Fig. 3. FTIR Spectra of G, T/G, CF/G, CF and T.

reagents (Kenya, packed in Egypt), respectively. Titanium dioxide (anatase) was purchased from Loba Chemie laboratory reagents (India). Sodium alginate (medium viscosity, about 2000 cps for 2% w/v aqueous solution at 25 °C, and average molecular weight of about 420 kDa), and $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ were purchased from Oxford laboratory reagents (India) and Nice laboratory reagents (India), respectively. $\text{FeCl}_3 \cdot 9\text{H}_2\text{O}$, $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ and As_2O_3 standard solutions were purchased from Alpha Chemika (India), Oxford laboratory reagents (India) and Chem Lab NV (Belgium), respectively.

2.2. Methods

2.2.1. Synthesis of nanofillers

The cobalt ferrite nanoparticles (CF) were synthesized by the common co-precipitation method [31]. Titanate nanotubes were synthesized by the hydrothermal method [32].

2.2.2. Synthesis of alginate microparticles (G)

The alginate microparticles were synthesized by the ionotropic gelation method as previously reported [33]. Briefly, the aqueous solution of sodium alginate (3% w/v) was slowly dropped into a solution of 100 ml CaCl_2 (2% w/v) with gentle stirring (300 rpm). Then, the obtained microparticles were left overnight in the CaCl_2 solution to strengthen. The strengthened microparticles were separated from the solution, washed with distilled water, dried overnight at 30 °C, and stored in sealed vessels until further investigation.

2.2.3. Synthesis of alginate nanocomposites

Alginate nanocomposite microparticles were obtained according to a previously reported method [34]. For the preparation of the CF/G nanocomposite, the CF nanoparticles were mixed with a sodium alginate solution (3% w/v) at a weight ratio of 1:3 (CF:G) with stirring vigorously for 3 h, followed by a slowly dropping of this homogeneous solution into CaCl_2 solution (2% w/v). The obtained nanocomposite particles were left overnight in the CaCl_2 solution to strengthen, then separated from the solution, washed with distilled water, dried overnight at 30 °C, and stored until further investigation. The same procedure was applied to prepare T/G nanocomposites.

2.2.4. Characterization

FESEM micrographs were taken by Quanta FEG 250 (Switzerland). HRTEM micrographs were obtained from JEOL-

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