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Granulation of Fe–Al–Ce nano-adsorbent for fluoride removal from drinking water by spray coating on sand in a fluidized bed

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ABSTRACT

A technology for the granulation of Fe–Al–Ce nano-adsorbent (Fe–Al–Ce) in a fluidized bed was developed. The coating reagent, a mixture of Fe–Al–Ce and a polymer latex, was sprayed onto sand in a fluidized bed. The granule morphology, coating layer thickness, granule stability in water and adsorption capacity for fluoride was investigated by analyzing samples for different coating time. The coating amount was from 3% to 36%. With increasing coating amount, granule stability decreased and adsorption capacity increased. FTIR analysis showed that the latex can react with active hydroxyl on the Fe–Al–Ce adsorbent, which led to a decrease of the adsorption capacity. Coated granules with a coating amount of 27.5% had a fluoride adsorption capacity of 2.22 mg/g (coated granules) at pH 7 and initial fluoride concentration of 0.001 M. A column test showed that 300 bed volumes can be treated with the effluent under 1.0 mg/L at an initial fluoride concentration of 5.5 mg/L, space velocity of 5 h^{-1} and pH of 5.8. The coating granulation of the Fe–Al–Ce adsorbent can produce granules that can be used in a packed bed for the removal of fluoride from drinking water.

1. Introduction

Excess fluoride in drinking water causes harmful effects such as dental and skeletal fluorosis [1]. The guideline values for fluoride in drinking water are 1.5 mg/L by the World Health Organization and 1.0 mg/L by China [2]. Adsorption is considered a more efficient technology for fluoride removal from drinking water when compared with other technologies like reverse osmosis, nanofiltration, electrodialysis and Donnan dialysis [2].

Activated alumina is the most widely used adsorbent because it is readily available and inexpensive. However, the need for frequent regeneration due to its low adsorption capacity at neutral pH results in complex operations [3], and the easy dissolution of aluminum in treated water leads to a secondary pollution. Bone char also can be used as a fluoride adsorbent, but it has a lower mechanical strength than activated alumina and it shows a weaker resistance to hydraulic shock in a packed bed [4].

A newly synthesized Fe–Al–Ce trimetal hydroxide adsorbent (Fe–Al–Ce) was reported to have a high adsorption capacity [2]. However, the Fe–Al–Ce adsorbent is available only as a fine powder or prepared in aqueous suspension as a hydroxide floc. In such forms, the adsorbent is limited to use in reactor configurations with large sedimentation basins or a filtration unit. Under such condition, the solid/liquid separation is fairly difficult. Besides, the Fe–Al–Ce adsorbent alone is not suitable as a filter medium because of its low hydraulic conductivity [5]. Therefore, its powder granulation to give granules of high strength is necessary so that it can be used in a packed bed.

Recently, researchers have developed the technique of coating an adsorbent onto sand to overcome the problem of using adsorbent powders in water treatment processes. Iron oxide-coated sand (IOCS) has been tested for removing cations and anions from synthetic and actual wastes [5]. The IOCS was prepared by the impregnation of sand in a mixed solution of salt and precipitator and subsequent drying [6,7]. However, the thickness of the coated layer was only several micrometers, which resulted in a low adsorption capacity. Furthermore, the coated layer can be easily shedded off, which left the coated sand with little adsorption capacity, and caused secondary pollution in drinking water.

In this paper, a Fe–Al–Ce adsorbent with an acrylic-styrene copolymer latex as a binder was spray-coated onto sand in a fluidized bed. The introduction of latex increased the stability of the coated layer [8]. The relationship between the coating properties and the adsorption properties, and the interaction between latex and Fe–Al–Ce adsorbent was investigated. A new type of granule adsorbent with

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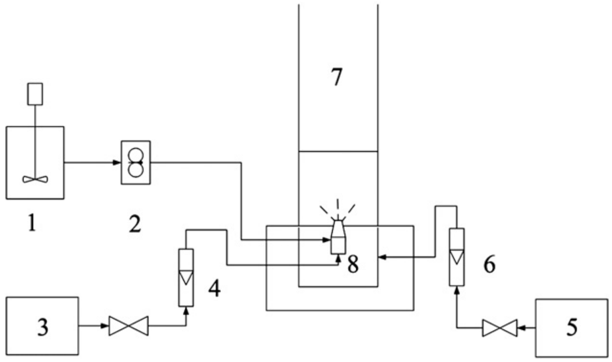

Fig. 1. Coating granulation apparatus. 1. Coating reagent vessel; 2. Peristaltic pump; 3. Atomized gas; 4. Flow meter; 5. Fluidized gas; 6. Flow meter; 7. Fluidized bed; 8. Nozzle.

high stability and adsorption capacity that is suitable for use in a packed bed was prepared.

2. Experimental

2.1. Materials

$FeSO_4 \cdot 7H_2O$, $Al_2(SO_4)_3 \cdot 12H_2O$ and $Ce(SO_4)_2 \cdot 4H_2O$ used were analytical grade (Chemical Engineering Company of Beijing, China). The other chemicals used were analytical reagents (AR) grade.

$FeSO_4 \cdot 7H_2O$, $Al_2(SO_4)_3 \cdot 12H_2O$ and $Ce(SO_4)_2 \cdot 4H_2O$ were dissolved in deionized water to form a mixed solution with concentrations of 0.1 M, 0.2 M and 0.1 M, respectively. 6 M NaOH solution was slowly added into the mixed solution until the pH was 9.5. The solution was stirred at 200 rpm during the whole process [2]. The precipitates obtained were centrifuged and washed with deionized water until the pH of the filtrate was 6.5 ± 0.2. The product, Fe–Al–Ce trimental hydroxide adsorbent (Fe–Al–Ce) with a diameter of 40 nm, was kept in deionized water.

Acrylic-styrene copolymer latex, which can crosslink and cure at room temperature, was supplied by the Institute of Polymer Science and Technology (Dept of Chem. Eng, Tsinghua University, China). This had a solid fraction of 40%, dynamic viscosity of 20 cP at 20 °C. The glass transformation temperature of the polymer latex was 22.8 °C.

The sand (Gaoyuan Stone Company, Beijing, China) was sieved to give the 1–2 mm fraction, soaked in HCl solution (pH = 1) for 4 h, rinsed with deionized water until the pH reached 6 ± 0.2, and dried at 105 °C for 24 h. The sand obtained was kept in capped bottles.

2.2. Coating method

A suspension of Fe–Al–Ce mixed with acrylic-styrene copolymer latex was used as the coating reagent. The mass ratio of Fe–Al–Ce to latex was 1:1 [8]. The latex was employed as a binder in the coating layer. The experimental apparatus for coating granulation is shown in Fig. 1. The reagent was agitated in a reactor to prevent the sedimentation of Fe–Al–Ce. The feed rate of the reagent was controlled by a peristaltic pump.

The sand was put into a fluidized bed with a diameter of 55 mm and a height of 500 mm. The sand was fluidized by controlling the gas velocity. The coating reagent was atomized and sprayed onto the sand, and the water in the coating reagent was dried by controlling the temperature of the fluidizing gas at 35 °C. After the spray coating was completed, the coated sand was kept in capped bottles.

2.3. Characterization

The coating amount was characterized by the mass ratio of Fe–Al–Ce to sand. The acrylic-styrene copolymer latex decomposed at 390 °C and burned at 450 °C according to the TG analysis. Thus, the coating amount can be determined by burning the granules in a muffle furnace and using an acid treatment. A known mass of granules was burned in a muffle furnace at 550 °C till the latex was burned off, to give burned sand (m_1). Then the burned sand was soaked in 1 M HCl solution for 3 h with agitation at 160 rpm until the coated layer was

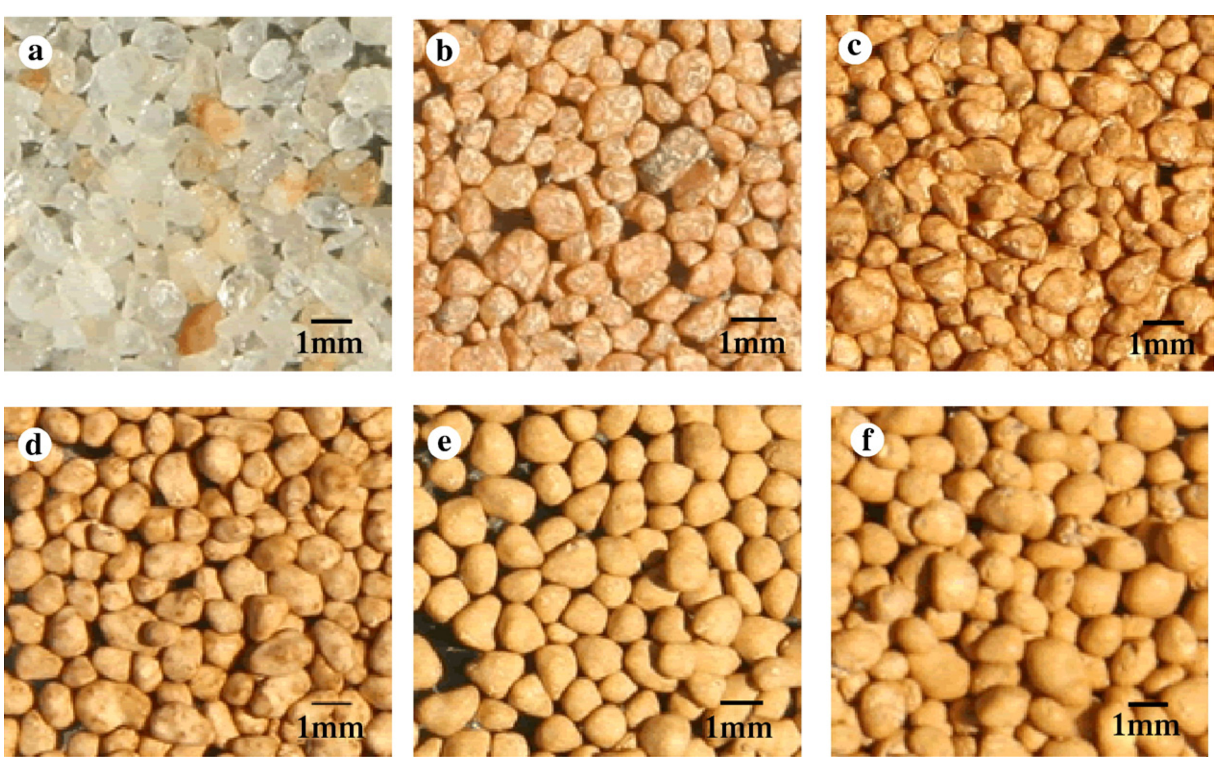

Fig. 2. Images of the granules during the coating process. Coating amount, %: a, 0; b, 3.55; c, 9.55; d, 18.40; e, 27.50; f, 36.20.