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Efficiency of barium removal from radioactive waste water using the combination of maghemite and titania nanoparticles in PVA and alginate beads



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HIGHLIGHTS

 \bullet TiO_2 and $\gamma\text{-Fe}_2\text{O}_3$ synthesized by hydrothermal and coprecipitation method respectively.

• Mag-tit PVA-alginate beads were used for barium ion removal from waste water.

• 9% of cesium ion was removed within 150 min at pH 8 under sunlight.

• The barium ion reduction fitted the pseudo second-order rate model.

• Mag-tit PVA-alginate beads can be reused for at least 7 times.

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ABSTRACT

In this paper, both maghemite (γ -Fe₂O₃) and titanium oxide (TiO₂) nanoparticles were synthesized and mixed in various ratios and embedded in PVA and alginate beads. Batch sorption experiments were applied for removal of barium ions from aqueous solution under sunlight using the beads. The process has been investigated as a function of pH, contact time, temperature, initial barium ion concentration and TiO₂: γ -Fe₂O₃ ratios (1:10, 1:60 and 1). The recycling attributes of these beads were also considered. Furthermore, the results revealed that 99% of the Ba(II) was eliminated in 150 min at pH 8 under sunlight. Also, the maghemite and titania PVA–alginate beads can be readily isolated from the aqueous solution after the process and reused for at least 7 times without significant losses of their initial properties. The reduction of Ba(II) with maghemite and titania PVA–alginate beads fitted the pseudo first order and second order Langmuir–Hinshelwood (L–H) kinetic model.

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1. Introduction

Adsorption

Barium is one of the most toxic elements present in radioactive liquid wastes due to long-lived radionuclides (e.g., 10.54 years for ¹³³Ba) (Roy et al., 2002) that can be generated by different activities such as: insufficient activities for the management and disposal of radioactive waste, accidental discharge of radioactive chemicals into the environment, nuclear practices, nuclear fuel cycle processes, etc. (Mishra and Singh, 1995a, 1995b; Mishra and Tiwary, 1999).

Some barium compounds that are released during industrial

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http://dx.doi.org/10.1016/j.apradiso.2015.06.028 0969-8043/© 2015 Published by Elsevier Ltd. processes dissolve easily in water and can be found in lakes, rivers, and streams. Due to their water-solubility these barium compounds can spread over great distances. When aquatic organisms such as fish, absorb the barium compounds, barium will accumulate in their bodies (Torab-Mostaedi et al., 2011). Because it forms insoluble salts with other common components of the environment, such as carbonate and sulfate, barium is not mobile and poses little risk. However, small amounts of water-soluble barium may cause some important diseases including: breathing difficulties, increased blood pressures, heart rhythm changes, etc. (Ghaemi et al., 2011).

There are a few studies on barium as a water pollutant but there are many reports on the behavior of other heavy metals pollutants such as lead, zinc, nickel, copper and mercury. Barium ought to be considered as a water pollutant because this element can be presented in drinking water and may cause health problems. The maximum amount of barium that is recommended by the United States Environmental Protection Agency (US EPA) is 2 ppm. Literature reviews from 1983 to 2009 reported that the concentration of barium in both groundwater and drinking water in several regions around the world varied from 0.001 to 6.4 mg/L. Thus, the removal of barium from water is in some of these places were necessary (Baeza-Alvarado and Olguín, 2011).

Since the advent of nuclear industries, various approaches and technologies were developed and adopted for the disposal and immobilization of radioactive aqueous wastes generated at different stages of the nuclear fuel cycle. The treatment process based on adsorption/ion exchange phenomenon plays an important role in preconcentration/separation of toxic radionuclides from aqueous wastes. A variety of sorbents/ion exchangers find their application in the treatment of diverse types of radioactive aqueous wastes (Stecher and Kogut, 1999; Mishra and Tiwary, 1999).

Recently, the correlation between titania (TiO₂) and wide band gap semiconductor has attracted many researchers attention due to its big pores and surface area that are essential in luminescence, photocatalysis, self-cleaning surfaces, highly specific chemical sensors, solar cells and carbon nanotubes (Brioude et al., 2004; Gao et al., 2009; Mital and Manoj, 2011; Soejima et al., 2013). Remarkably, one of the most popular photocatalysts is TiO₂ because of its great thermo stability and photocatalytic effectiveness. Recently, considerable research has been conducted on the use of TiO₂ powders in photodegrading different organic pollutants in the water. Additionally, the studies on how to enhance the catalytic attributes of TiO₂ have also considerably increased. A number of researchers have begun to couple semiconductor materials, such as TiO₂/CdS (Jia et al., 2007), ZnO/CdS (Castillo et al., 2001) and SnO₂/Fe₂O₃ (Xia et al., 2008) so as to expand its general application (Wang and Gui, 2013).

On the other hand, Fe₂O₃ with band-gap of 2.2 eV is an appropriate candidate and a remarkable n-type semiconducting material to be utilized as a photocatalyst in sunlight irradiation (Idris et al., 2013). The Fe₂O₃photocatalytic nature is studied in water splitting (Kiwi and Gratzel, 1987), semiconductor electrode functions and photo degradation of organic contaminants (Chatterjee et al., 1994). Furthermore, Fe₂O₃ can be applied as a sensitizer to TiO₂photocatalyst because of its narrow band-gap. As Fe_2O_3 -TiO₂ is exposed with sunlight; the electrons in the Fe_2O_3 valence group can easily attack the conduction group and make holes. Employing the construction of the built-in field in Fe₂O₃-TiO₂ heterojunction causes the electrons in the valence bands of TiO₂ to transmit into Fe₂O₃ since the photo created holes transmit the TiO₂ valence band in the opposite direction. An efficient procedure in improving the photocatalytic movement of the composition is the control transmission between the valance bands of TiO₂ and Fe₂O₃, since it leads to an increase in the electron-hole recombination time(Akhavan and Azimirad, 2009).

Some studies used alginate to encapsulate magnetic nanoparticles to produce it in the form of beads. Polyvinyl alcohol (PVA) is another polymer that is commonly used to remove heavy metals of waste water because it is non-toxic, safe and cheap. But PVA has tendency to agglomerate, therefore PVA is usually combined with alginate to reduce the agglomeration (Idris et al., 2008).

Previous studies (Idris et al., 2010, 2012a, 2012b) have showed that γ -Fe₂O₃ nanoparticles in PVA–alginate beads have been used as adsorbents to remove Pb(II). Themajority of prior research also utilized γ -Fe₂O₃ PVA–alginate beads, which is formed by utilizing γ -Fe₂O₃ nanoparticles in PVA and alginate beads while considering wastewater polluted with Cr(VI) (Idris et al., 2012a, 2012b), Cd(II) (Idris and Majidnia, 2014a, 201b) and Cs(I) (Majidnia and Idris, 2015). However studies on the use of combined TiO₂ nanoparticles and γ -Fe₂O₃ nanoparticles embedded in PVA alginate in treating barium ions has yet to be explored. Thus in this research an effort is made to improve the performance of barium ions removal by using photo catalyst beads containing both TiO₂ and γ -Fe₂O₃ nanoparticles embedded in the PVA–alginate beads.

2. Materials and methods

2.1. Materials

Iron(II) chloride, iron(III) chloride, trisodium citrate, sodium alginate, acetone, boric acid, nitric acid, ammonia solution, acetylacetone, hydrochloric acid, polyvinyl alcohol (PVA), calcium chloride, titanium isopropoxide, urea and Ba(II) solution were bought from Sigma-Aldrich. Additionally, ethanol was bought from Merck.

2.2. Methods

2.2.1. TiO_2 powder synthesis

Initially 2 ml titanium isopropoxide and 2 ml acetyl acetone was added to 40 ml ethanol solution and stirred continuously at room temperature. In another beaker, 0.5 g of soluble urea was dissolved in 10 ml of deionized water. Both of these solutions were then added in drops to 40 ml of deionized (DI) water with continuous stirring until a pale yellow solution is formed with pH 5.6. The solution was kept under vigorous stirring for one hour and then transferred into a 120 ml Teflon lined stainless steel autoclave which was then placed in an oven and heated at 150 °C for 18 h. The content was finally cooled to room temperature. The yellowish white colored sample was washed with ethanol and deionised water and then centrifuged. The sample was then dried at 80 °C for 3 h and then stored in a clean dark colored dry bottle for further use (Thapa et al., 2012).

2.2.2. Preparation of γ -Fe₂O₃ nanoparticles

The maghemite $(\gamma$ -Fe₂O₃) nanoparticles were produced through co precipitation of a stoichiometric combination of ferric and ferrous chlorides in an ammonium hydroxide solution (Bee et al., 1995). The initially acquired magnetite (Fe₃O₄) precipitate was acidified via nitric acid and oxidized into maghemite (γ -Fe₂O₃) at 90 °C with iron(III) nitrate. Additionally, the particles were coated by citrate anions in order to achieve an unchanging maghemite distribution, which is well matched with an alginate gel as a neutral medium. Subsequently, the coated nanoparticles were distributed in water after precipitation with acetone to get a constant ferro fluid with pH 8. The maghemite nanoparticles characteristics were defined in our prior paper (Idris and Majidnia, 2014; Idris et al., 2011, 2010).

2.2.3. Preparation maghemite and titania PVA-alginatebeads

100 mL of precursor solution was prepared using 12 g of PVA and 1 g of alginate in presence of deionized water. After which 12 g of PVA was dissolved using 72 mL of distilled water and heated at 80 °C for 5 h. The PVA was also heated in a microwave for 4 min to attain dissolution. Meanwhile, alginate was dissolved in 20 mL of deionized water in another beaker and then the PVA and alginate solution were mixed together.

- i. Preparation maghemite PVA–alginate beads: 0.1 g of γ -Fe₂O₃ was added to above solution.
- ii. Preparation titania PVA–alginate beads: 0.1 g of TiO_2 was added to above solution.
- iii. Preparation maghemite and titania PVA–alginate: γ-Fe₂O₃ nanoparticles were mixed with TiO₂ nanoparticles in different

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