Chemosphere 200 (2018) 405–411

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Fe-Ti/Fe (II)-loading on ceramic filter materials for residual chlorine removal from drinking water

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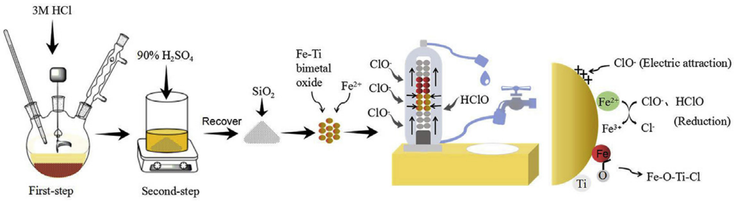
HIGHLIGHTS

- Recycle of SiO_2 from red mud as raw material is environmental friendly.
- The ceramic filter material was modified by Fe (II) and Fe-Ti bimetal oxide.
- The modified ceramic filter material has good removal effect on residual chlorine.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 16 September 2017
Received in revised form
18 February 2018
Accepted 25 February 2018
Available online 26 February 2018

Handling Editor: Xiangru Zhang

Keywords:
Red mud
Fe (II)/Fe-Ti bimetal oxide
Residual chlorine removal
Hydroxyl group

ABSTRACT

Ceramic filter material was prepared with silicon dioxide (SiO_2), which was recovered from red mud and then modified with Fe (II) and Fe-Ti bimetal oxide. Ceramic filter material can be used to reduce the content of residual chlorine from drinking water. The results showed that after a two-step leaching process with 3 M hydrochloric acid (HCl) and 90% sulfuric acid (H_2SO_4), the recovery of SiO_2 exceeded 80%. Fe (II)/Fe-Ti bimetal oxide, with a high adsorption capacity of residual chlorine, was prepared using a 3:1 M ratio of Fe/Ti and a concentration of 0.4 mol/L Fe^{2+}.

According to the zeta-potential, scanning electron microscopy (SEM) and Brunauer–Emmett–Teller (BET) analysis, Fe (II) and Fe-Ti bimetal oxide altered the zeta potential and structural properties of the ceramic filter material. There was a synergistic interaction between Fe and Ti in which Fe–O–Ti bonds on the material surface and hydroxyl groups provided the active sites for adsorption. Through a redox reaction, Fe (II) transfers hypochlorite to chloride, and Fe–O–Ti–Cl bonds were formed after adsorption.

1. Introduction

In urban water supply systems, chlorine disinfection in water has been widely adopted (Cai et al., 2017; Zhang et al., 2009). In order to restrain the propagation of pathogenic bacteria, viruses and other pathogenic organisms, a proper quantity of residual chlorine should be retained in the water to persistently kill microorganisms (Sorlini and Collivignarelli, 2005). Therefore, keeping a certain concentration of chlorine in drinking water is necessary (Gómez-López et al., 2014). Residual chlorine is the main compound in chlorinated drinking water that is, suspected to be carcinogenic (Helbling and Vanbriesen, 2008; Ren et al., 2009). It has been reported that free residual chlorine is present mainly in the form of the hypochlorite ion and hypochlorite in the water, and surplus residual chlorine can affect people's taste and destroy the quality of water (Jović et al., 2015). The WHO has provided a guideline limitation of free chloride in pipe networks of between 0.03 mg/L and 0.4 mg/L (WHO, 2011).

Various methods such as activated carbon adsorption and redox processes utilizing KDF filter medium have been investigated for

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https://doi.org/10.1016/j.chemosphere.2018.02.156

residual chlorine removal (Filtr, 2008). Nevertheless, some limitations were presented during the water treatment process, such as high cost, high doses and low efficiency of materials (Zeng et al., 2012). Among these methods, adsorption is considered to be more efficient for residual chlorine removal from drinking water because it is simple to operate and cost-effective.

Iron oxides and hydroxides are gaining popularity due to their high capacity for chlorine removal (Wang et al., 2015). Similar to iron, titanium is one of the most prevalent elements in the environment. Titanium dioxide is becoming one of the most attractive environmental materials due to its nontoxicity and chemical stability (Wajima et al., 2009; Ishihara et al., 2002). It was reported that a new chemical bond, formed between the two metal elements through an oxygen atom increased the number of hydroxyl groups on the adsorbent surface and the adsorption capacity of the materials (Yu et al., 2016). Therefore, it can be anticipated that if the titanium oxide were doped into the ferric oxide, the composite metal oxides would not only retain the advantages of their single component oxides but may also be able to significantly improve their sorption capacity. However, due to their tiny size, nanosized metal oxide adsorbents are hard to separate from water, which could cause secondary pollution for drinking water (Yang et al., 2012). Red mud is the by-product of alumina production through the Bayer chemical process, and it is a complex material whose chemical and mineralogical composition varies widely (Davris et al., 2016). Many researchers have attempted to develop techniques for recovering its valuable elements (Borra et al., 2016; Wang et al., 2013). According to recent research, red mud contains rich stores of iron and silicon. SiO_2 is the main component, and it is the main material utilized for ceramic filter materials (Yang et al., 2015). Therefore, recovering valuable components from red mud for making granular material is a sustainable method of red mud disposal and could allow for easy separation of ceramic material from water (Ghosh et al., 2011).

In this paper, SiO_2 was the major component leached from red mud, which was then used as the main material to prepare ceramic filter material. The raw samples were modified by Fe (II) and Fe-Ti bimetal oxide. A series of Fe (II) and Fe-Ti bimetal oxides with different Fe/Ti molar ratios were synthesized to investigate the effect of the surface properties of ceramic filter material on residual chlorine removal. In addition, the reaction mechanism of residual chlorine with Fe (II)/Fe-Ti bimetal oxide was studied.

2. Material and methods

2.1. Chemicals and materials

The red mud was obtained from China Shandong Aluminum Industry Company, and ferric chloride hexahydrate ($FeCl_3 \cdot 6H_2O$) was recovered from the red mud. Sodium carbonate, limestone, polyvinyl alcohol (PVA), bentonite, titanyl sulfate ($TiOSO_4$) and hydrochloric acid (HCl) were analytical grade and were obtained from Tianjin Kermel Reagent Co. All solutions were prepared with deionized water. The chemical composition of the red mud is listed in Table S1. The X-ray diffraction (XRD) patterns of the red mud are shown in Fig. S1. It was evident that the three major phases were quartz (SiO_2), hematite (Fe_2O_3) and gibbsite ($Al(OH)_3$) which exist in air-dried red mud.

2.2. Sample pretreatment

The preliminary experiments were performed according to the schematic procedure as shown in Fig. 1. The red mud leaching procedure was carried out in a round-bottom split reactor with three-necks, and 0.5 g of the red mud was added to the reactor. According to the chemical activity difference in the metal ions in the red mud, the red mud was leached by different concentrations and different types of acid step by step, to separate the metal ions. After the first leaching step with 3 M HCl, the leaching ratio of Fe was 95.4%, which was recovered in the form of $FeCl_3 \cdot 6H_2O$. After the second leaching step with 90% H_2SO_4, the recovery of SiO_2 exceeded 80%, which was the main material used to prepare ceramic filter materials.

2.3. Modified sample preparation

The ceramic filter material was prepared with SiO_2 as the main raw material, which was extracted from red mud. Bentonite, limestone, pulverized coal, and sodium carbonate were injected as adminicular materials. All of the raw materials were sieved through a 200 mesh screen and then mixed to a the mass ratio of 65:25:4:4:2. After the powder was evenly blended, 2 g of mixed powder was added to 2 ml of polyvinyl alcohol (5%), and the mixture was churned continuously until becoming a viscous form at 75 °C. Then, an aperture board was used to squeeze the viscous form before shredding it to a diameter of 1.0–2.0 mm, which was

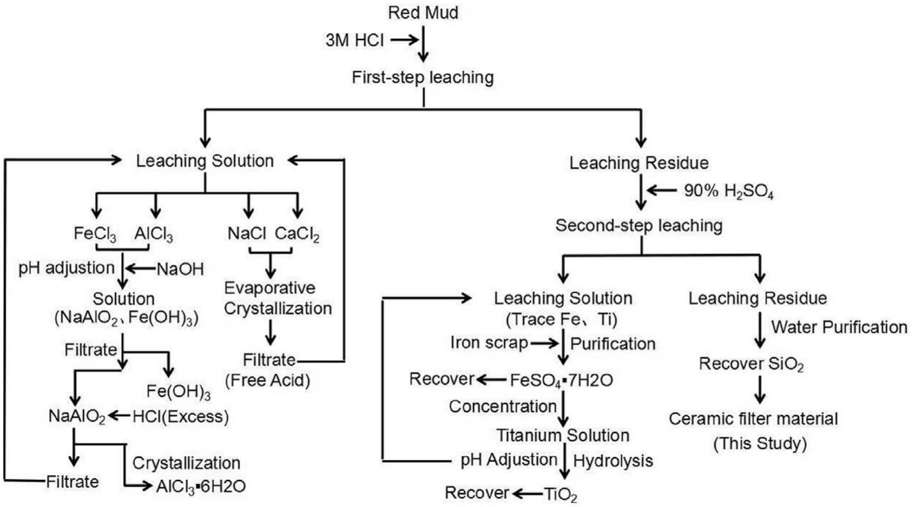

Fig. 1. The procedure for preparation of ceramic filter material from red mud.