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The adsorptive removal of chromium (VI) in aqueous solution by novel natural zeolite based hollow fibre ceramic membrane

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A R T I C L E I N F O

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A B S T R A C T

Adsorption is one of the most efficient ways to remove heavy metal from wastewater. In this study, the adsorptive removal of hexavalent chromium, Cr (VI) from aqueous solution was investigated using natural zeolite, clinoptilolite, in the form of hollow fibre ceramic membrane (HFCM). The HFCM sample was prepared using phase inversion-based extrusion technique and followed by sintering process at different sintering temperatures in the range of 900–1050 °C. The fabricated HFCM was characterised using scanning electron microscopy (SEM), contact angle, water permeability, and mechanical strength for all HFCMs sintered at different temperatures. The adsorption and filtration test of Cr (VI) were performed using an in-house water permeation set up with a dead-end cross-flow permeation test. An asymmetric structure with sponge- and finger-like structures across the cross-section of HFCM was observed using SEM. Based on the characterisation data, 1050 °C was chosen to be the best sintering temperature as the water permeability and mechanical strength of this HFCM were 29.14 $L/m^2{\cdot}h$ and 50.92 MPa, respectively. The performance of the HFCM in adsorption/filtration was 44% of Cr (VI) removal at the Cr (VI) concentration of 40 mg/L and pH 4. In addition, the mathematical model was also performed in simulating the experimental data obtained from this study. All in all, the natural zeolite-based HFCM has a potential as a single-step Cr (VI) removal by membrane adsorption for the wastewater treatment.

1. Introduction

With the rapid growth of the global industrialisation, many problems related to the environment are emerging, which creates risks not only to the environment, but also to the ecological systems as well as human health. Nowadays, heavy metals are becoming among the most hazardous pollutants in the supply and treated water and are imposing a great harm to human health and aquatic lives due to the ability of bioaccumulation in body and its high toxicity (Miretzky and Cirelli, 2010; Sarin and Pant, 2006). Various adverse effects related to the mental and neurological system could be resulted by the exposure, transportation, and accumulation of the heavy metals such as mercury, lead, arsenic, and cadmium (Dong et al., 2017; Rehman et al., 2017). Of all of these heavy metals, chromium is known as one of the top sixteen major toxic contaminants that has detrimental effects on human health (Gardea-Torresdey et al., 2000).

Chromium exists in aqueous media in two prominent oxidation state forms namely trivalent Cr (III) and hexavalent Cr (VI) ions and these are the most abundantly found forms of chromium in water body. Naturally, chromium is found in the environment in the form of Cr (III). However, the oxidation of this type of chromium have generated the Cr (VI) ions during various industrial processes. This industrial waste species could be toxic and can be accumulated in many physical and chemical components in the environment. Cr (III) is said to be less toxic than Cr (VI), and have a significant role and necessary for lipid and sugar metabolism, making it as an essential trace element for human and animal (Bai et al., 2015; Prasad, 2013). In addition, it is advisable for a human to take Cr (III) of about 50–200 $\mu g{\cdot}day^{-1}$ (Herrero-Latorre et al., 2018). Although this species is less toxic to human as well as essential for dietary nutrient, the excess quantities of Cr (III) may cause damages to aquatic organism and would eventually disrupt the food chain. Therefore, the permitted concentration of these Cr species in the surface or potable water must be less than 0.05 mg/L (Baral and Engelken, 2002). Meanwhile, Cr (VI) is more poisonous and is known to

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be carcinogenic and mutagenic in nature (Rojas et al., 2005). Cr (VI) is known to be 100 times more toxic than Cr (III) due to its high water mobility and solubility, and also because it can be easily reduced. The carcinogenic effect of Cr (VI) can be strong by causing failure of major organs such as kidney and liver as well as dermatitis and diarrhoea (Mohan and Pittman Jr, 2006; Mohan et al., 2005). According to the World Health Organization (WHO), the permissible level of chromium in surface water bodies is 0.05 mg/L. However, its concentration in industrial wastewater ranges from 0.5 to 270 mg/L (Ghosh, 2009). In addition, the chromium level in water may vary depending on the geographical and standard limits set by the law of a country. According to the Environmental Protection Agency (EPA) and the European Union (EU), the permitted level of Cr in water must be lower than 0.05 mg/L (Baral and Engelken, 2002). The Indian standard of the maximum contaminant level (MCL) of the total chromium contained in water is set as 0.05 mg/L (Pradhan et al., 2017). For the Malaysian standards, the allowable concentration of Cr has been limited to 0.05–0.1 mg/L, regulated by the Malaysian Food Regulations (Azlan et al., 2012). Therefore, the elimination of Cr (VI) becomes a vital issue as the permissible amount in the wastewater discharge becomes more stringent.

Many conventional treatments have been developed to remove Cr (VI) from water bodies, including ion exchange (Rengaraj et al., 2001), nanoparticles (Gupta et al., 2016), electrocoagulation (Akbal and Camcı, 2011), and adsorption/filtration (Babel and Kurniawan, 2003). For all the methods, the capital and operational costs normally set limitations in the efficiency f the Cr (VI) removal from the wastewater. In addition, the production of secondary wastes as well as the formation of a large quantity of slug has further reduced the effectiveness of these methods (Babel and Kurniawan, 2003; Hegazi, 2013). Thus, the searching of low-cost and yet effective method in the Cr (VI) removal is in a real demand and needs immediate attention. One of the promising ways of contaminant removal from wastewater is by the means of membrane separation technique.

Recently, the membrane technology has become an excellent way for wastewater treatment due to its ability of not only separating the contaminants from the water bodies, but also purifying the treated water. The semipermeable property of the membranes is known to be the special feature that only permits several elements or compounds to pass through them. This special feature of porous membrane is applicable for many purposes such as wastewater treatment (Abbasi et al., 2010), drinking water purification (Bellobono et al., 2005), water desalination (Fang et al., 2012), gas separation (Nafisi and Hagg, 2014), and bacteria removal (Kroll et al., 2010). However, only a limited number of studies reported on the treatment of wastewater containing heavy metals via membrane technology.

Most of the commercially available membranes are made of polymeric materials. The low cost and mass production of polymer have made the polymeric membrane widely used in the industries. Polymers that are commonly used to fabricate polymeric membranes are polyvinylidene fluoride (PVDF), polysulfone (PSf), polyethersulfone (PES), and polyacrylonitrile (PAN) (Kong and Li, 1999; Melbiah et al., 2017; Ochoa et al., 2003; Zhang et al., 2017). However, these membranes are functioning effectively only in the mild conditions. There are also some other disadvantages such as inability to separate volatile compounds and the tendency to be fouled quickly, which will eventually lower the flux of the membrane and thus, reduce the performance. Therefore, the attention now is moving towards the potential of the ceramic membrane as an alternative to replace the polymeric membrane.

Over the past few years, many researchers have been working on the development of ceramic membrane with low cost and yet high performance for various applications. The superior properties of the ceramic membrane over the polymeric membrane have made the ceramic membrane more suitable for many applications. The exceptional resistance to high temperature and pressure, low fouling effect, and high endurance towards strong cleaning agents are among the advantages offered by the ceramic membrane (Bader, 2007; Basumatary et al., 2016; Xu et al., 2014). Despite all these advantages, the usage of the ceramic membrane is always redundant due to high production cost. This could be attributed to the expensive raw materials such as alumina, zirconia, titania, and silica. In addition, most of these materials require high sintering temperature to reach a compromise between porosity, permeability, and mechanical strength (Liu et al., 2003, 2016). This in turn could upsurge the production outlays of the membranes (Hubadillah et al., 2017a, 2018). Therefore, finding the alternative of a low cost and highly performing membrane is of utmost urgency.

Zeolite is a hydrated aluminosilicate mineral naturally found in nature. Clinoptilolite is the most abundant source of natural zeolite found across the globe, making it among the cheapest natural zeolite sources in the market. This mineral is composed of symmetrically stacked alumina and silica tetrahedra and forms an open and stable three-dimensional structure with a negative charge (Burgess et al., 2003). This negative charge allows for the adsorption of certain positively charged ions. Thus, this feature has made the natural zeolite a potential heavy metal cations adsorbent. On another note, the adsorption process normally requires the contact between the adsorbate and adsorbent for a specific time. In this process the adsorbents are normally suspended in the adsorbate solution in powder form. However, this conventional method is favourable since the used adsorbent needs to be separated and further cleaned for reuse (regeneration), which may cause the loss of adsorbent particles. For that reason, a single step of adsorption and separation should be invented to prolong the life span of the adsorbent.

The fabrication of the zeolite-based hollow fibre ceramic membrane (HFCM), which possesses both features of adsorption and separation, is the main aim of this study. The hybrid process combining both adsorption of Cr (VI) and the separation of the adsorbed species by filtration from the feed in a single step is the novelty of this study. The effect of the HFCM fabrication parameter, namely sintering technique, on the physical properties and the performances of the membranes was studied throughout this work. The cheap raw material (natural zeolite) and the low sintering temperature will eventually produce an inexpensive HFCM that possesses a promising adsorption capacity in the removal of the Cr (VI) from the wastewater.

2. Materials and methods

2.1. Materials

The natural zeolite (clinoptilolite) powder (with an average particle size of 70 μm) was purchased from Liaoning Province, China (Shijiazhuang Mining Trade Co. Ltd.). Polyethersulfone, PESf (Radal A300, Ameco Performance, USA), N-methyl-2-pyrrolidone, NMP (AR grade, Qrec™) and Arlacel P135 (polyethylene glycol 30-dipolyhydroxystearate, Uniqema) were used as the polymer binder, dispersant, and solvent, respectively. The fabrication of the zeolite-based hollow fibre was performed using the phase inversion-based extrusion technique followed by sintering.

2.2. Hollow fibre ceramic membrane (HFCM)

2.2.1. Fabrication of hollow fibre membrane

Prior to the dope suspension preparation, the pre-ground natural zeolite powder was sieved with a 36 μm sieve and was dried overnight at 60 °C to remove all the trapped moisture. The suspension composition of the zeolite-based HFCM (45 wt.%) was 45 g of zeolite powder, 1 g of Arlacel P135 as the dispersant, 5 g of PESf as the polymer binder, and 49 g of NMP as solvent. The HFCM spinning parameters were 10 mL/min, 15 mL/min, and 5 cm for the suspension extrusion rate, bore fluid flow rate, and air gap distance, respectively.

The zeolite-based HFCM was fabricated via the phase inversion-based extrusion and sintering technique. The preparation of the dope