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The effect of thermal activation for natural zeolite on the performance of ceramic membrane as Pb^{2+} ion adsorption

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Abstract. Zeolites known as molecular filters have the potential to adsorb heavy metals in wastewater, but zeolites in nature have many impurities, so it was activated to remove impurities and increase the effectiveness of zeolite adsorption. This work was conducted to determine the performance of thermally activated ceramic membrane as a heavy metal filter for wastewater. The ceramics made with the wet-mixing method are activated zeolite powder mixed with polyethylene glycol (PEG) solution which is useful as an adhesive as well as a pore-making agent. The fabrication was done of each with a PEG fraction (w/w) of 8.33%; 10.93%; 13.23%; 15.27%; 17.1%; and 18.75%, respectively. The flow test was performed using Pb solution as wastewater with concentration 200 ppm, and it was obtained ceramic permeability between of $1x10^{-11}$ thus $6x10^{-11} m^2$, while UV-Vis spectroscopy was done to obtain the filter performance (C/C_0), that is 0.13; 0.20; 0.32; 0.36; 0.45; 0.54. In conclusion zeolite membrane thermally activated can be used as heavy metals filter.

1. Introduction

Rapid populating growth and inefficiencies in progressing waste have the potential to damage nature. Wastewater is a type of water that can cause serious problems, the water contaminated by metals like lead (Pb), iron (Fe), cadmium (Cd), can damage river ecosystem such as through the food chain[1-4]. It's vital to filter the water to remove the metals ions in water so that the water becomes feasible for nature [5].

Lead is one of the potentially harmful heavy metals when adsorbed into the body [6]. The lead is non-biodegradable and can accumulate in living tissues, therefore becomes concentrated throughout the food chain and can be easily absorbed into the human body [7]. Exposure to lead in drinking water even at low concentration level may associated with a wide range of effects, including various neurodevelopmental effects, mortality (mainly due to cardiovascular diseases), impaired renal function, hypertension, reduced fertility, adverse pregnancy outcomes [8], anemia, encephalopathy, hepatitis and nephritic syndrome [9]. Lead toxicity in humans is well-known; it replaces calcium and, consequently, can accumulate in the skeletal system [10]. Lead is widely used in many essential industrial applications such as storage battery manufacturing, printing, pigments, fuels, photographic materials, explosives industry, pulp and paper, petrochemicals, refineries, ceramic manufacturing, glass fabrication, paint, oil, metal, phosphate fertilizer, electronic goods, wood production, combustion of fossil fuel, forest fires, mining activity, automobile emissions, sewage wastewater and sea spray [11-13].

Various chemical and physicochemical methods for the treatment of wastewaters comprising lead pollutant are known, such as chemical precipitation, electrochemical, ion exchange, biosorption and adsorption [14-18]. However, most of these methods may be highly expensive or ineffective when the metals are dissolved in large volumes of a solution at a relatively low concentration [19]. Adsorption is very often used for wastewater treatment because it is ordinarily preferred for lead removal due to its easy handling, high efficiency, availability of different adsorbents and cost-effectiveness. The main properties of these adsorbents are strong affinity and high loading capacity [20].

Today, membrane separation processes are taken into consideration as essential techniques for adsorption of wastewater treatment and have found large applications in various fields. Each membrane separation process uses a membrane, thereby selectively separates one component from the other. This separation is due to the distinguish in physical and chemical properties of the membrane and the permeable element. As ceramic membranes are receiving much more attention nowadays, they are often compared to polymeric membranes. One of the main advantages of the ceramic membrane is having a capability to endure extremely aggressive environments, such as highly acidic solution [21,22].

Ceramics membrane is one material which potential for filter application [4]. Having mesoporous size in both particles and pores makes ceramics can filter metal ions in wastewater [23]. In this work used natural material and inorganic types with the aim is low cost, easy to fabricate, and products that can be used repeatedly [24]. Zeolites known as molecular filters have the potential to adsorb heavy metals in wastewater, but zeolites in nature have many impurities, so it was activated to remove impurities and increase the effectiveness of zeolite adsorption [25,26].

Using membrane ceramic as filters have been studied by Masturi *et al* 2012 used clay, and Aji *et al.* 2015 used glass waste with permeability $0.3 \times 10^{-15}\mathrm{m}^2 - 25 \times 10^{-15}\mathrm{m}^2$ and $1.49 \times 10^{-16}\mathrm{m}^2 - 4{,}50 \times 10^{-15}\ \mathrm{m}^2$ respectively [24,27]. The permeability value is relatively low, so a method is needed to increase the permeability value. Furthermore, this study obtained a permeability value of $1.31 \times 10^{-11} - 6{,}75 \times 10^{-11}\ \mathrm{m}^2$, we get high permeability due to the considerable use of the pore-making fraction. Therefore, the addition of fractions from the pore-making material on the ceramic was carried out while still maintaining filtering effectiveness [4]. This work was conducted to determine the performance of zeolite thermally activated ceramic membrane as a heavy metal filter for wastewater, with Pb^{2+} ion in solution as test material.

2. Methods

2.1. Material

Natural zeolite as main material purchased at Pandanaran (West Java, Indonesia) and Polyethylene Glycol (PEG) 4000 as poring agent purchased at Bratachem (Indonesia).

2.2. Pretreatment and thermally activated of natural zeolite

Natural zeolites were powdered by blending for 10 minutes and sieved using 100 mesh sieve. The natural zeolite powder was thermally activated using the microwave at a temperature of 105^{o}C for 24 hours. The amount of natural zeolite powder must be as minimal as possible with the vessel to determine to prevail thermally activated.

2.3. Fabrication of zeolite ceramic membrane

The fabrication was done using a wet mixing method. PEG 4000 powder 25 g dissolved in 25 mL of distilled water and further stirred using magnetic stirrer for 2 hours, then natural zeolite thermally activated were mixing with varying fraction PEG solution (8.33; 10.93; 13.23; 15.27; 17.1; 18.75). Furthermore, the mixture was heated at 100^{o}C for 35 minutes, finally to obtained zeolite ceramics membrane, the mixture was pressed at 5000 psi for 10 minutes and fired using furnace at 1000^{o}C for 8 hours [28].

2.4. Pressing method for fabrication of zeolite ceramic membrane

The ceramic membrane can be fabricated using different methods depending on the application requirements, the desired membrane structure, and the specific materials. The most common manufacturing processes are pressing method. Pressing method is a well-known method used mainly for the fabrication of ceramic membranes for fundamental research. This method is commonly based on the pressing of a dry powder utilizing a press machine. Indeed, after mixing the powder homogeneously (raw material with pore-forming agent ratios), the obtained product is pressed uniaxially (Fig. 1), that is, it undergoes stress by a punch in a mold with immobile walls to achieve the desired membrane support shape. This process allows very high production rates. For consolidation, the obtained flat membrane support must go through heat treatment, generally at a temperature reaching that of the used materials sintering [29].

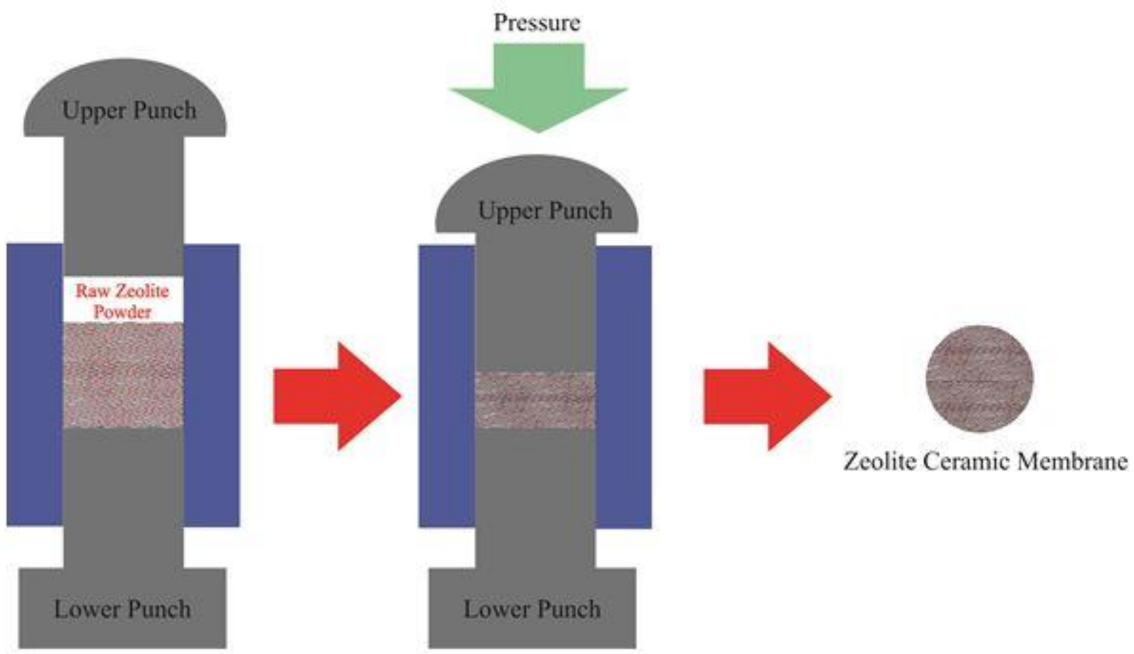

Figure 1. The instrument of pressing method for fabrication of zeolite ceramic membrane

2.5. Performance of zeolite ceramic membrane

Performance of ceramics membrane was done using test fluid, and the permeability was known using Darcy's law [4]. The test solution was prepared by dissolving $PbCO_3$ 0,02 g in water which then filtered through the membrane. The UV-Vis absorption spectra characteristics of the membranes were measured using Lambda 25 Perkin Elmer spectrometer. Lead content in pre-filtration and post-filtration of test fluid was determined heavy metal, the UV-Vis spectra around a wavelength of 214 nm were examined [30]. The performance of ceramics was defined as the ratio of Pb^{2+} ion remaining in the filtrate to the initial Pb^{2+} ion concentration in the test fluid [24].

3. Results and Discussion

The permeability ceramics membrane after flow test is shown in Fig. 2. The permeability value of ceramic was increased depending on the PEG content. Higher PEG content was making increase pores and expand the surface area of the membrane. As a result, make time faster when Pb^{2+} ion in solution past through the ceramic membrane and the permeability increased [31,32]. Furthermore, thermally activation influence structure zeolite hence its high permeability value with range 10^{-11} m^2 [33].

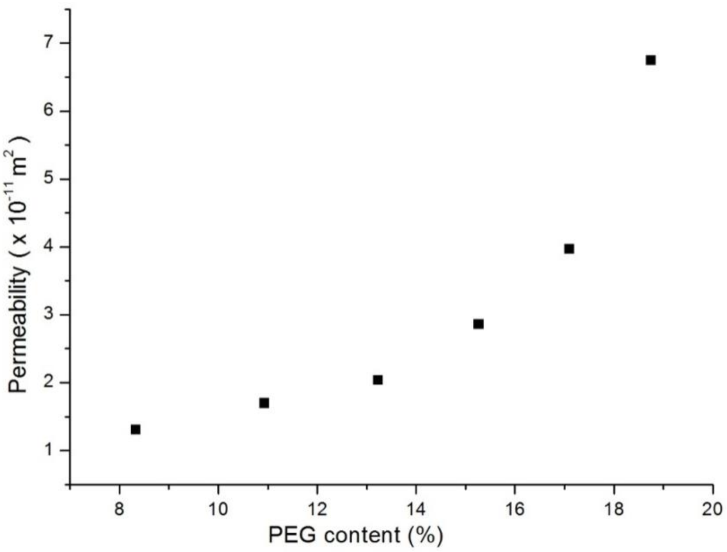

Figure 2. Zeolite ceramic membrane's permeability against PEG content

Thus, the principle is that the higher the PEG content, the higher the porosity of the ceramic. Hence there will be lower Pb^{2+} ion adsorbed by pores' wall. There is inverse proportionality relation between adsorbed solute concentration with a value of permeability, where more the adsorbed solute when the smaller the value of the permeability of the ceramic membrane. Fig. 3 is shown the relation between permeability and filter performance C/C_0, which is a ratio of solute in the solution post-fluid test to initial concentration pre-test.

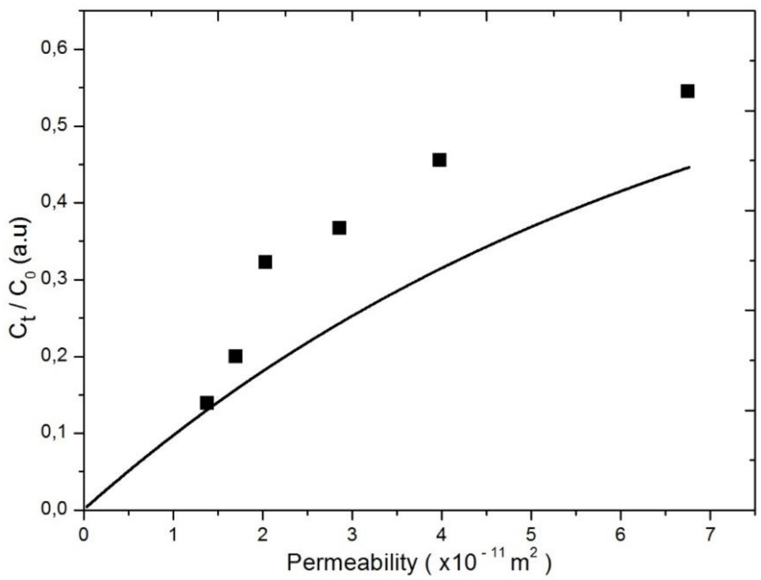

Figure 3. The relation between permeability and filter performance C/C_0 on zeolite ceramic membrane

Consider molecular interaction between Pb^{2+} ion with pore's wall. Some of the ions will at some point reached a pore of a size such that only water molecule may pass through. This may happen with zeolite framework nanopore with a scale of 3-5 times of Pb^{2+} ionic radius (1.33 Å) [34]. Pb^{2+} ion cannot pass through the nanopores even with its smaller size due to the overall attractive force exerted by zeolite nanopore's wall on Pb^{2+} ion [35]. Suppose the decrease concentration is proportional with the initial concentration and at minimal permeability, there is no Pb^{2+} ion passing the membrane.

4. Conclusion

The ceramics made with the wet-mixing method are activated zeolite powder mixed with polyethylene glycol (PEG) solution. The fabrication was done of each with a PEG fraction (w/w) of 8.33%; 10.93%; 13.23%; 15.27%; 17.1%; and 18.75% respectively.The permeability was measured using a method based on Darcy's law. In membrane with maximum PEG fraction and test fluid's Pb concentration of 200 ppm, fabricated filter exhibit values of permeability between 1.31×10^{-11} thus 6.75×10^{-11}m^2 and filter was able to reduce the lead concentration in test solution by 87% with detail filter performance (C/C_0), that is 0.13; 0.20; 0.32; 0.36; 0.45; 0.54. In conclusion zeolite membrane thermally activated can be used as heavy metals filter.

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