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Evaluation of iron and manganese-coated pumice from Sungai Pasak, West Sumatera, Indonesia for the removal of Fe (II) and Mn (II) from aqueous solutions

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Abstract

The objective of this study was to evaluate the performances of iron and manganese-coated pumice from Sungai Pasak, West Sumatera, Indonesia as the adsorbents for removal of Fe (II) and Mn (II) from a queous solutions. The effect of soaking time for iron and manganese coating was evaluated and as comparison, the adsorption of Fe (II) and Mn (II) using uncoated pumice was conducted. The experiments were performed in batch mode at room temperature (20-25°C), pH 7; 10 g/L of adsorbent dose; 0.30-0.4 mm of adsorbent diameters; 100 rpm of agitation speed and 90 minutes of contact time. In addition, the desorption process of Fe (II) and Mn (II) from the three kinds of adsorbent was also investigated. The results showed that the optimum soaking time for iron and manganese coating for removal of Fe (II) and Mn (II) were 100 and 48 hours, respectively. Iron-coated pumice showed to have high removal efficiencies of two ions compared to uncoated and manganese-coated pumice. More than 84% of Fe (II) and 72% of Mn (II) with initial concentration of 15 and 5 mg/L, respectively, were removed by 10 g/L iron-coated pumice, while by using uncoated and manganese-coated pumice, the removal efficiencies were less than 75% for the two ions. The desorption study noticed that up to 20% of Fe (II) and 100% of Mn (II) were recovered from the three kinds of pumice adsorbent. Overall study indicated that pumice from Sungai Pasak may be a promising adsorbent for iron and manganese removal from water and wastewater.

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1. Introduction

Iron and manganese, which being second most abundant metal in the earth's crust, are commonly present in ground water supplies used by many water systems. The presence of iron and manganese in the drinking water is not harmful to human bodies. However, higher concentration causes discoloration, staining, turbidity and bad taste problems. It also forms iron oxide or manganese dioxide accumulations in pipes. Therefore, World Health Organization has approved the treatment of water if concentrations of iron and manganese are higher than 0.3 mg/L and 0.1 mg/L. 1.2.3

Several techniques have been applied to remove metals, like iron and manganese from groundwater including chemical precipitation, ion exchange, electrodialysis, adsorption process, membrane separations, reverse osmosis, and solvent extraction. Among these methods, adsorption is a most common technique for the metal removal because adsorption is low cost, has a simple design, is easy to perform and is insensitive to toxic substances ⁴. Adsorption onto activated carbon has been widely applied for removing metals from water and wastewater. However, adsorbent-grade activated carbon is expensive, and the regeneration of activated carbon for reuse increases the cost ^{5,6}. Therefore, more interests have recently arisen in the investigation of low-cost adsorbents with a good sorption capacity to remove metal ions from water and wastewater.

For the past few decades, several researchers have reported on the potential use of agricultural byproducts as good adsorbents for the removal of metal ions from aqueous solutions and wastewaters, such as pine bark wastes ⁷, rice husk ash ⁸ and maize husk ⁹. On the other hand, the use of natural geomaterials or local minerals such as zeolite ^{10.11}, better that a such as adsorbents for metal removal also have been widely considered in recent years.

Pumice is a natural pozzolan produced by release of gases during cooling and solidification of lava. Porous structure of pumical is created by the formation of bubbles or air voids when gases in molten lava are trapped during cooling. The mice has a large surface area, skeleton structure and low weight, where it normally either floats on water 14,15. Pumice mainly has been used for structural applications such as aggregate in light weight concrete, cements, and filters. Nowacal s, pumice also has been used as adsorbent for pollutant removal from water and wastewater 16. However, the direct usage of unmodified mineral adsorbents, including pumice may bring several problems, such as 1 w adsorption capacity. Therefore, these materials must be treated before application in adsorption process. Modification of pumice by cationic surfactants increased anionic contaminant retention, hydrophobicity and cation exchange capacity 13,17.

In this study, the capability of pumice from 3 ungai Pasak, West Sumatera, Indonesia, after being coated with iron and manganese, as adsorbent for removal of Fe (II) and Mn (II) from aqueous solutions was evaluated. This local mineral is available in a great abundance, as byproduct of the process of sand mining in that area. For this 4 dy, the effect of soaking time for iron and manganese coating was determined and as comparison, 4 adsorption of Fe (II) and Mn (II) using uncoated pumice was also conducted. Moreover, the desorption process of Fe (II) and Mn (II) from the three kinds of adsorbent was investigated as well.

2. Materials and methods

2.1. Reagents

All used chemicals in this study were reagent grade from Merck (Germany). Fe (II) and Mn (II) stock solutions were prepared by dissolving of ammonium iron (II) sulfate (Fe (NH₄)₂ (SO₄)₂.62₂O) and manganese (II) sulfate (MnSO₄.4H₂O) in distilled water. Either NaOH or HNO₃ were used for adjusting pH of the solutions to the desired values.

2.2. Preparation of adsorbents

Pumice stone was collected at the verside of Sungai Pasak, West Sumatera, Indonesia as byproduct of the cocess of sand mining in that area. Before coating Fe and Mn on pumice surface, the stone was crushed and sieved in order

to produce particle size fractions of 0.30-0.50 mm. The obtained particles were immersed in 37% HCl for 24 h and were washed several times using distilled water. To prepare iron and manganese-coated pumice, solutions of 0.5 M Fe (NH₄)₂ (SO₄)₂.6H₂O and MnSO₄.4H₂O were used to soak the particles of pumice and the pH of solutions were adjusted to 12 and 8, respectively, by adding NaOH. The effect of soaking time was studied by varying the time of 24 h, 48 h, 72 h and 100 2 The beakers containing soaking particles of pumice were placed in a static state at room temperature (20-25°C) and then dried in the oven at 110°C for 24 h. Finally, the dried particles were washed three 2 hes by distilled water and then oven dried again at 110°C for 24 p. In Incoated pumice was prepared by washing three times by distilled water and then oven dried at 110°C for 24 h. A scanning electron microscopy (SEM, model S-3400N, Hitachi, Japan) was used to observe the surface of coated and uncoated pumice.

2.3. Batch adsorption experiments

Batch adsorption experiment was carried out at room temperature (20-25°C), pH 7; 10 g/L of adsorbent dose; 0.30-0.50 mm of adsorbent diameters; 100 rpm of agitation speed and 90 minutes of contact time. In each experiment, 100 mL of Fe (II) and Mn (II) solutions of known initial concentration were treated with three kinds of adsorbents in a set of erlenmeyer flasks and shaken with a shaker machine at a speed of 100 rpm. After 90 minutes of contact time, the adsorbent 3 vere separated from the metal solutions by centrifugation at 2500 rpm for 5 min. The supernatants were measured for the concentration of Fe (II) and Mn (II) using atomic absorption spectrophotometer (Rayleigh WFX 320, China). The amount of metal ions adsorbed by the pumice was gotten as the difference between the initial and final ion concentrations of the solutions. All experiments were repeated three times and results presented are, consequently, the averaged values of replicate tests.

The removal efficiency, R (%), was calculated using the equation (1), below:

$$R = \frac{c_0 - c_e}{c_e} \times 100\% \tag{1}$$

where C_0 and C_e are the initial and equilibrium concentrations of metals (mg/L), respectively.

2.4. Desorption experiment

Coated and uncoated pumices used in the adsorption experiment, after being separated from the solution, were subjected to desorption experiments using 0.1 N HCl. The adsorbents were placed in several flasks containing 0.1 N HCl and shaken at room temperature (20-25°C) and 100 rpm for 90 min. Furthermore, the suspensions were centrifuged at 2500 rpm for 5 min. The supernatants were measured for metal ion concentrations using atomic absorption spectrometry (Rayleigh WFX 320, China).

The percent of desorption was obtained from the following equation (2)

$$Percent of desorption = \frac{Concentration of metal desorbed}{Concentration of metal adsorbed} \times 100\%$$
 (2)

1 3. Results and discussion

3.1. Effect of soaking time in iron and manganese coating

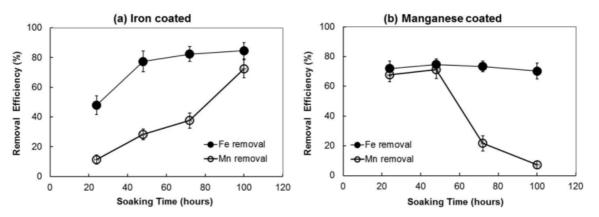


Fig 1. The effect of soaking time of iron (a) and manganese (b) coating on Fe (II) and Mn (II) removal [at room temperature (20-25°C), pH 7; 10 g/L of adsorbent dose; 0.30-0.50 mm of adsorbent diameters; 100 rpm of agitation speed; contact time: 90 min, initial metal concentrations: 15 mg/L for Fe (II) and 5 mg/L for Mn (II)].

Effect of soaking time is one of important parameter in iron and manganese coating of panice for adsorption process. The effect of 24 h, 48 h, 72 h and 400 h of soaking time in iron and manganese coating on Fe (II) and Mn (II) removal is shown in Figure 1(a) and (b). The initial concentrations of Fe (II) and Mn (II) were 10 mg 3 and 5 mg/L, respectively. Figure 1(a) shows the effect of soaking time for iron coating on removal efficiencies of Fe (II) and Mn (II) from aqueous solutions. An increasing trend with increasing soaking time was observed for removal of both ions and the highest removal efficiencies were found at 100 h of soaking time with 84 % and 72% for Fe (II) and Mn (II), respectively. Same increasing trend of removal efficiencies by increasing of soaking time was also observed in manganese coating of pumice. However, the different 4 sults were obtained in manganese coating of pumice, as shown in Figure 1(b). The highest removal efficiencies of Fe (II) and Mn (II) were found at 48 h of soaking time. At that time, the removal efficiencies of Fe (II) and Mn (II) reached to 74 % and 71%. It shows decreasing removal efficiency with increasing soaking time with a steep drop up to 48 h soaking time.

The results indicated that different types of metals have different optimum soaking time for pumice coating. Prior to reaching the optimum soaking time, the maximum surface coating may not be achieved on the pumice that affect their capabilities in adsorption metal ions. By the optimum soaking time, iron and manganese coating on pumice was expected to increase the adsorption capacity of pumice by increase their surface area and pore structure ¹⁹. Furthermore, 100 h soaking time fo on coating and 48 h for manganese coating were applied to pumice, which were subjected to adsorption process for removal of Fe (II) and Mn (II) from aqueous solutions.

3.2. Adsorption of Fe (II) and Mn (II) onto the coated and uncoated pumice

After being coated by iron and manganese, pumice was used as adsorbent in the adsorption of [3] (II) and Mn (II) from aqueous solutions. To define the apacity of pumice, both coated and uncoated pumice were applied in the adsorption process. Figure 2 displays the removal efficiencies of Fe (II) and Mn (II) from aqueous solutions. Iron-coated pumice showed to have [4] th removal efficiencies of Fe (II) and Mn (II) compared to uncoated and manganese-coated pumice. More than 84% of Fe (II) and 72% of Mn (II) with initial concentration of 15 and 5 mg/L, respectively, were removed by 10 g/L iron-coated pumice, while by using uncoated and manganese-coated pumice, the removal efficiencies were less than 75% for the two ions.

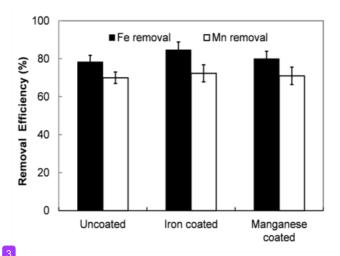


Fig 2..Removal efficiencies of Fe (II) and Mn (II) from aqueous solutions by using three kinds of adsorbent: uncoated, iron and manganese coated [at room temperature (20-25°C), pH 7; 10 g/L of adsorbent dose; 0.30-0.50 mm of adsorbent diameters; 100 rpm of agitation speed; contact time: 90 min, initial metal concentrations: 15 mg/L for Fe (II) and 5 mg/L for Mn (II)].

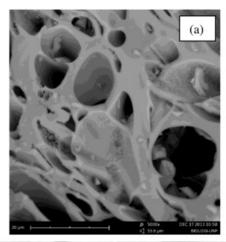
Some researchers were reported that iron coating increased the surface areas of adsorbent, such as in iron oxide coated sand ^{20,21,22}, iron oxide coated olivine ²⁰ and iron coated pumice as well ^{18,19}. Increasin 5 he surface areas by iron coating might have occurred through modification of pore structures and sizes as well as the attachment of iron oxides with much higher areas.

The SEM images of the uncoated and coated pumice from Sungai Pasak showing surface morpholog of the samples at magnification of 5000x are illustrated in **Figure 3**. The surface of the uncoated pumice showed a highly porous, smooth surface, cellular and irregular texture with larger cavities, which provides suitable sites for adsorption (**Figure 3a**). After being coated, pumice Sungai Pasak showed the different pore structure, particularly for iron coated pumice. As seen in **Figure 3b**, the pore morphological of pumice changed from 5 homogeneous to heterogeneous structure, may be due to the decrease in particle structure. The iron coated pumice are filled and partially blocked with iron oxides and due to coating, the morphology of iron and manganese coated pumices were appeared to be porous and rough with defined cavities. However, the extent of porosity and rough surface of iron coated was greater than that of manganese coated (**Figure 3c**). The present observation is in agreement with the research studies conducted by Kitis et al. (2007), Far et al. (2012), Sepen al. (2014), Shayesteh et al. (2016)

On the other hand, for it 5 coated pumice, iron oxide coatings mask or overwhelm the surface electric 5 properties of the underlying material. Iron coating generally decreased the amounts of total surface basic groups. In addition to impacts on surface areas and pore structures, iron oxide species bound on pumice particles also modify original surface chemistries. Such factors may significantly affect the reactivity of original pumices, and they should be considered when pumice particles are used as a support material, adsorbent, or filtration media 19.

3.3. Desorption study

Desorption experiment was performed to elucidate the mechanism of metal ion removal and recovery from the three kinds pumice adsorbent; uncoated, iron coated and manganese coated. The data in **Figure 4** gives the percent desorption of the Fe (II) and Mn (II) from the three kinds pumice adsorbent. It showed that at the end of 90 min contact time, it was noticed that up to 20% of Fe (II) and 100% of Mn (II) were recovered from the three kinds of pumice adsorbent.



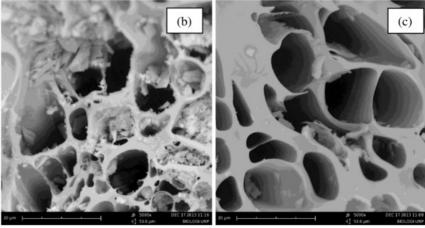
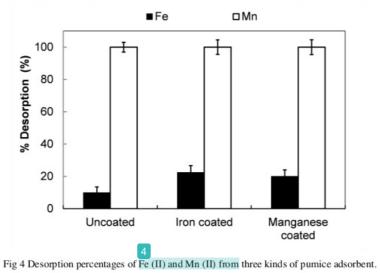


Fig 3. SEM images of uncoated pumice (a), iron coated pumice (b) and manganese coated pumice (c).



These results indicated that the same interaction may occur between Fe (II) and Mn (II) as adsorbate and the three kinds of pumice as adsorbent. Comparatively, desorption of both metal ions from the pumic adsorbent is greater for Mn (II) than Fe (II) in all kinds of adsorbent studied. This observation may be due to the formation of stronger metal-pumice bond with Fe (II) as a result of smaller ionic radius. Ionic radius is the distance between the nucleus and the electron in the outermost shell of an ion. The smaller ionic radius, the stronger bond between metal ion and pumice, the more difficult to release from the adsorbent ²³.

4. Conclusions

In this study, the potential of local mineral, pumice from Sungai Pasak, West Sumatera, Indonesia, has been aluated for the adsorption of Fe (II) and Mn (II) from aqueous solutions. The SEM images of this pumice showed a highly porous, smooth surface, cellular and irregular texture with larger cavities, which provides suitable sites for adsorption. Modification the pumice by coating with iron and manganese can increase the adsorption capacity of pumice by increase the surface area of the adsorbent through modification of pore structures and sizes as well as the attachment of iron oxides with much higher areas. Soaking for 100 h and 48 h in coating process were applied to obtain iron and manganese coated pumice, respectively, which has given the highest removal efficiency for both metals. Iron-coated pumice showed to have high removal efficiencies of two ions with more than 84% of Fe (II) and 72% of Mn (II) at the initial concentration of 15 and 5 mg/L, respectively. At the end of 90 min contact time, it was found that up to 20% of Fe (II) and 100% of Mn (II) were desorbed from the three kinds of pumice adsorbent indicated that the same interaction may occur be oven Fe (II) and Mn (II) as adsorbate and the three kinds of pumice as adsorbent. These results indicated that pumice from Sungai Pasak, West Sumatera, Indonesia would be used as a potential and alternative adsorbent for the removal of Fe (II) and Mn (II) from water and wastewater.

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