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Preparation and Application of Fly Ash-Based Geopolymer for Heavy Metal Removal

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Abstract. Geopolymer is anorganic polymer which can be used to heavy metal removal through adsorption process. Geopolymer can be produced from raw material containing alumino-silicate oxide such as fly ash which is solid waste from coal combustion. In this study, geopolymer was prepared from fly ash with alkali activation process using mixture of 10 N NaOH solution and Na-Silicate solution. Fly ash-based geopolymer was then applied as adsorbent for different heavy metals removal, i.e. Cu^{2+}, Fe^{2+}, Mn^{2+}, Zn^{2+}, in aqueous solutions. Removal efficiencies were calculated from changes of heavy metal concentration determined by Atomic Absorption Spectroscopy (AAS) analysis. Characterization of fly ash-based geopolymer covered Brunauer-Emmett-Teller (BET) surface area analysis and Scanning Electron Microscope-Energy Dispersive X-Ray (SEM-EDX) analysis. The results showed that fly ash-based geopolymer could adsorb heavy metals with better removal efficiency compared to the raw fly ash.

INTRODUCTION

Heavy metal pollution has become one of the serious environmental problems. Heavy metals are materials that cannot be decomposed and in certain levels can endanger the health of humans and other living things. The presence of heavy metals in the environment can naturally occur through the process of weathering minerals, erosion, or volcanic activity; as well as from human activities such as mining, metal processing, use of pesticides and fertilizers [1]. Treatment of heavy metal wastewater can be carried out by physical methods (adsorption, membrane filtration, coagulation and flocculation, flotation), chemical methods (precipitation, ion exchange, electrochemical), and biological methods (bio-adsorbent, use of microorganisms) [2]. Among these methods, adsorption is widely used because the process is simple, effective, and low cost. Heavy metal adsorbents that have been widely used are activated carbon, zeolite, resin, fly ash, chitosan, etc. [3].

Geopolymer, inorganic polymer with Si-O-Al polymeric bonds having amorphous to semi-crystalline structure, can also be used as heavy metal adsorbents. Geopolymer can be made from materials containing alumino-silicate oxides with alkaline activators at temperatures below 100 ^oC [4]. Fly ash, a solid waste from coal combustion, contains silica and alumina which can be utilized as raw material for geopolymer. Several studies have shown that geopolymer with fly ash as raw material has been used as heavy metal adsorbent, such as copper [5], cadmium [6], chromium [7], and lead [8].

This paper studied the preparation of geopolymer from fly ash and application of geopolymer as adsorbent for heavy metal removal, namely Cu^{2+}, Fe^{2+}, Mn^{2+}, Zn^{2+}, in aqueous solutions. Changes of heavy metal concentration were determined by Atomic Absorption Spectroscopy (AAS) analysis, meanwhile fly ash-based geopolymer was characterized by Brunauer-Emmett-Teller (BET) surface area analysis and Scanning Electron Microscope-Energy Dispersive X-Ray (SEM-EDX) analysis. As comparison, the heavy metal adsorption process was also carried out with fly ash adsorbent.

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METHODS

Geopolymer Preparation

Fly ash used in this study was from power plants in East Java, Indonesia. Fly ash was sieved with 200 mesh standard sieve before used as geopolymer raw material. The main oxides content in fly ash based on X-Ray Fluorescence (XRF) analysis were SiO_2 (42.4%), Al_2O_3 (22.6%), CaO (14.8%), and Fe_2O_3 (9.4%). The content of silica, alumina, and iron oxide in fly ash reaches >70% so that it can be categorized as class F fly ash [9].

In the preparation of geopolymer, ratio of fly ash to alkaline activator was 2.5:1. Alkaline activator consisted of 10 N NaOH solution and Na-Silicate solution in a ratio of 1:1. Na-silicate solution had solid mass fraction of 0.35 and SiO_2/Na_2O mole ratio of 3.2. Fly ash was mixed with alkaline activator for 10 minutes. The mixture is then placed in a 5x5x5 cm mold. After 24 hours, the geopolymer was removed from the mold and heated in an oven at 60 ^oC for 6 hours. The geopolymer was cured for 7 days in room temperature before crushed and sieved with 50 mesh standard sieve. Geopolymer powder was then used as heavy metal adsorbent.

Adsorption Test

Solutions of Cu^{2+}, Fe^{2+}, Mn^{2+}, and Zn^{2+} with concentrations of 100 ppm were each made from analytical grade $CuSO_4.5H_2O$, $FeSO_4.7H_2O$, $MnSO_4.H_2O$, and $ZnSO_4.7H_2O$ with aquadest. A 0.2 g of geopolymer powder were added to a 100 ml of heavy metal solution and stirred for 2 hours. The adsorption process was carried out at pH 5 and room temperature. After the adsorption process was complete, the geopolymer powder was separated from the solution by filtration. The concentration of heavy metal solutions was measured by Atomic Absorption Spectroscopy (AAS). The removal efficiency (%) was calculated from the equation:

$$Removal\ efficiency\ (\%) = \frac{C_0 - C_e}{C_0} x100 \quad (1)$$

C_0 is the initial heavy metal ion concentration, and C_e is the heavy metal ion concentration at equilibrium.

Characterization

Characterization was carried out on fly ash and geopolymers, comprising BET surface area analysis with Quantachrome Instruments and SEM-EDX analysis with JEOL JSM-6510LA instruments.

RESULTS AND DISCUSSION

Fly Ash-Based Geopolymer

Geopolymer had been prepared from fly ash with mixture of 10 N NaOH solution and Na-Silicate solution as alkaline activator. The alkaline activation process had changed fly ash which had spherical particles into a geopolymer which had an amorphous and porous structure as shown in Fig.1. Based on the results of BET analysis, fly ash that passed through 200 mesh standard sieve had surface area of 12.141 m^2/g, while fly ash-based geopolymer that passed through 50 mesh standard sieve had surface area of 27.205 m^2/g. The higher geopolymer surface area indicates that geopolymer can be used as adsorbent better than fly ash [10].

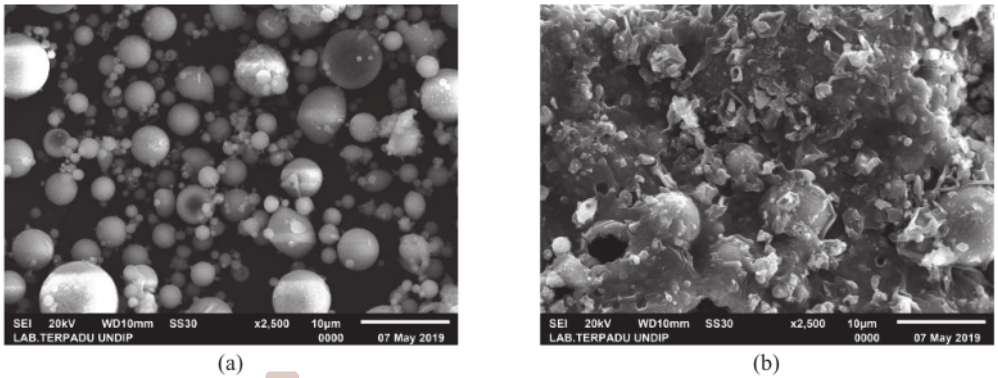

FIGURE 1. SEM micrographs of fly ash (a) and fly ash-based geopolymer (b).

Heavy Metal Removal

The efficiency of heavy metal removal by adsorption process using fly ash and fly ash-based geopolymer adsorbents is shown in Fig.1. The removal efficiency of heavy metals with fly ash-based geopolymer adsorbent was better than that with fly ash adsorbent. Besides fly ash-based geopolymer having larger surface area than fly ash, the presence of negative charges on Al in geopolymer will attract cations or heavy metal ions [11].

Adsorption of heavy metals in both fly ash-based geopolymer and fly ash had the same tendency, namely Cu^{2+} > Fe^{2+} > Zn^{2+} > Mn^{2+}. Based on the ion size, Mn^{2+} ion has the largest size, followed by Fe^{2+}, Cu^{2+}, and Zn^{2+} ions so that Mn^{2+} ions were the least absorbed. Zn^{2+} ion has the smallest size, but Zn^{2+} ions were less adsorbed than Cu^{2+} and Fe^{2+} ions. This result is in accordance with the result obtained by Yücel [12] with fly ash adsorbent, namely Cu > Zn > Mn and Sahoo et al. [13] with alkaline modified fly ash adsorbent, namely Fe > Zn > Mn.

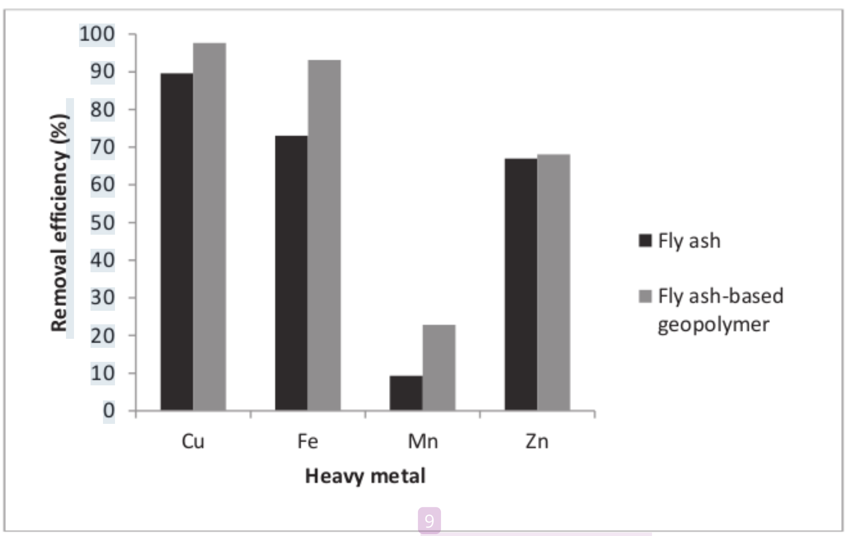

FIGURE 2. Heavy metal removal efficiency by fly ash and fly ash-based geopolymer adsorbents.

The microstructure of fly ash-based geopolymer after heavy metal adsorption process is shown by Fig.3. Heavy metal ions appeared to cover the surface of the geopolymer. The results were also supported by the results of SEM-EDX analysis as shown in Table 1. Geopolymer after adsorption of Cu^{2+} ions showed an increase in the Cu content compared to geopolymer before adsorption. This also occurred on geopolymer after adsorption of Fe^{2+}, Mn^{2+}, and Zn^{2+} ions. The increase of Cu content on the geopolymer surface was the highest compared to the increase of Fe, Mn, and Zn contents.

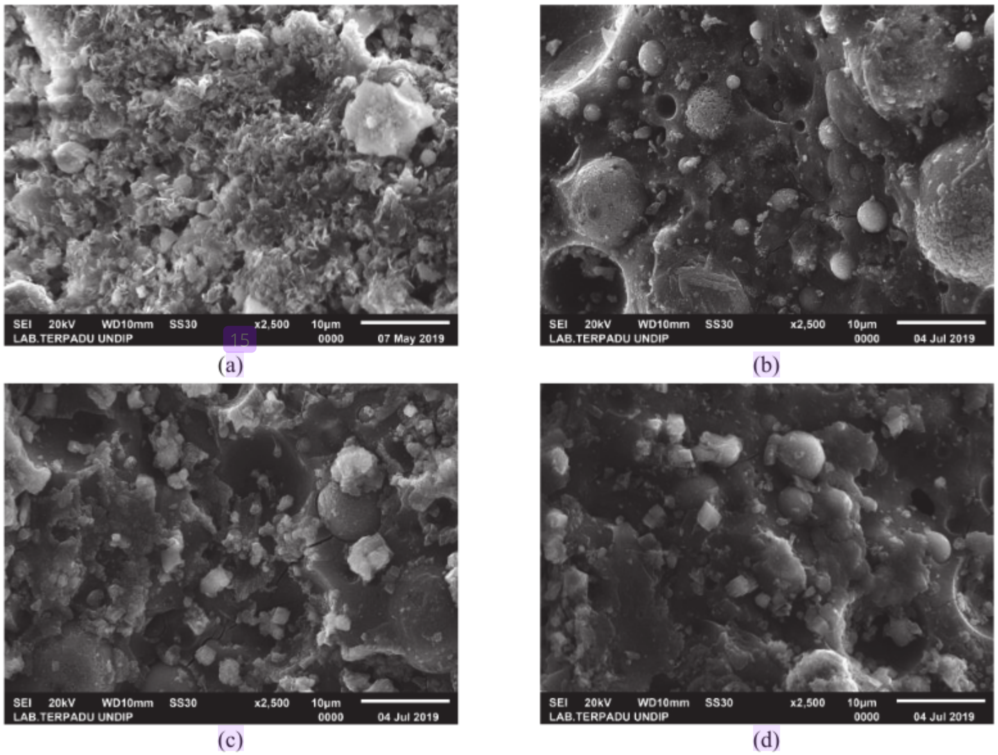

FIGURE 3. SEM micrographs of fly ash-based geopolymer after adsorption of Cu^{2+} (a), Fe^{2+} (b), Mn^{2+} (c), and Zn^{2+} (d) ions.

TABLE 1. Composition of elements measured via EDX on FAG (fly ash-based geopolymer before adsorption process), FAG-Cu (fly ash-based geopolymer after Cu adsorption process), FAG-Fe (fly ash-based geopolymer after Fe adsorption process), FAG-Mn (fly ash-based geopolymer after Mn adsorption process), and FAG Zn (fly ash-based geopolymer after Zn adsorption process).

Composition (%)	FAG	FAG-Cu	FAG-Fe	FAG-Mn	FAG-Zn
Al	20.86	19.34	24.40	21.86	19.58
Si	36.39	35.58	44.34	41.48	42.63
Na	22.9	2.08	4.19	3.44	5.36
Ca	13.18	25.15	12.53	19.14	21.68
Cu	0.27	12.48	0.93	0.57	0.20
Fe	6.11	5.35	11.01	6.38	8.94
Mn	0.07	0.00	0.15	6.78	0.05
Zn	0.22	0.00	2.45	0.34	1.57

CONCLUSIONS

Geopolymer had been prepared from fly ash with alkaline activator which was mixture of 10 N NaOH solution and Na-silicate solution. The adsorption sequence of heavy metal ions were $Cu^{2+} > Fe^{2+} > Mn^{2+} > Zn^{2+}$ both with fly ash-based geopolymer adsorbent and fly ash adsorbent. The application of geopolymer from fly ash as adsorbent of heavy metal ions (Cu^{2+}, Fe^{2+}, Mn^{2+}, Zn^{2+}) showed better removal efficiency than fly ash.

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