Development of Geopolymers Composite Based on Metakaolin-Nano ZnO for Antibacterial Application

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Abstract. A research on the development of geopolymers composite based on metakaolin-nano ZnO for antibacterial application has been conducted. The aim of this study was to investigate the influence of nano ZnO addition in the production of antibacterial composite. Nano ZnO was produced through precipitation of ZnSO₄ in NaOH solution. The composites were developed through alkali activation of metakaolin added with nano ZnO at various concentrations, namely 5%, 10%, & 15% relative to the weight of metakaolin. The structure and the phase of the starting and the resulting materials were examined by using x-ray diffraction (XRD). The resulting composites were immersed in water taken from a well for several days to investigate their antibacterial properties. The morphology and the presence of bacteria on the surface of composites were investigated by using scanning electron microscopy (SEM). It was found that geopolymers composites based on metakaolin-nano ZnO is an effective composite to limit the growth of the bacteria.

1. Introduction

Geopolymer is a material produced through geosynthesis of aluminosilicate minerals. The polymeric group of this material consist of SiO₄ and AlO₄ with a tetrahedral structure [3]. Geopolymers is synthesized through alkali activated of various aluminosilicate minerals such as metakaolin, fly ash, and furnace slag at temperature below [12]. Geopolymers has been known as a good binder material and therefore can be incorporated with other materials to form a new composite with excellent properties [14]. Basically, composite consists of a matrix and aggregate which act as reinforcement. In this study, geopolymers binder was produced by using metakaolin. Metakaolin is produced through dehydroxilation of kaolin at 750°C for 6 hours [13]. Metakaolin is considered as a standard material for geopolymers production due to its level of purity compared to other aluminosilicate materials such as fly ash. Antibacterial composites have been developed from various starting materials. One of the material which has excellent properties as antibacterial is Ag-nanoparticle (N-Ag) [2][5][11]. It is known that the size of Ag particle related directly to its ability as antimicrobial [1][6]. Besides N-Ag, ZnO nanoparticle has also been studied as antibacterial material [4]. ZnO is a promising material because of its good semiconducting properties, easy to produced, cheap, and environmentally friendly [10]. In this study, nano ZnO was used as an aggregate to produce geopolymers composite. The presence of nano ZnO will form a composite which has an antibacterial properties particularly if the size of ZnO particle is below 100 nm [8].
2. Experimental Method

ZnO nanoparticle was produced through precipitation of ZnSO4 particle in NaOH solution. The mixture was stirred for 3 hours until ZnO particle was produced. The particles was then washed three times with demineralized water and then with alcohol. The resulting particles was heated up to 110 °C for 4 hours until white nano ZnO was produced. Geopolymers binder was produced through alkali activation of metakaolin. ZnO nanoparticle was mixed with geopolymers binder at various concentrations; 5%, 10%, and 15% relative to the mass of metakaolin. The mixture was molded and cured at 70°C for 1 hour. The resulting materials were stored in open air for 2 weeks before conducting any measurements. The antibacterial test was performed by immersing the resulting composite into a water taken from a well for 3 days. The presence of any bacteria on the surface of the samples was examined by using Scanning Electron Microscope (SEM). The structure and chemical composition of the starting and the resulting materials were studied by using x-ray diffractions (XRD). The compressive strength measurement was performed to get information about the strength of the material due to the presence of nano ZnO.

3. Result and Discussion

The XRD of the resulting nano ZnO is shown in figure 1. It can be seen that the material is a single phase typical to ZnO mineral.

![Figure 1. Diffractogram of nano ZnO produced in this study](image)

The particle size of nano ZnO was calculated by using Scherrer equation for the highest peak in figure 1. It was found that the particle size of ZnO produced in this study was around 17 nm. This result indicates that the produced ZnO fulfill the criteria as nanoparticle [9].

Figure 2a shows the diffractogram of the as prepared composite, and figure 2b is the diffractogram of the same sample after immersed in water for several days.
Figure 2(a) shows the diffractogram of as prepared samples containing different percentage of nano ZnO. The diffractogram indicate the amorphous nature of geopolymers structure. The presence of nano ZnO start to appear at the addition of 10% and become discernible at 15%. Figure 2(b) shows the diffractogram of the samples after immersing into water for several days. It can be see that the peak of amorphous hump shifted into a smaller 2θ indicating rearrange of geopolymers structure. The peaks of ZnO at the sample with 10% nano ZnO disappear which is probably leach out during the immersion process. The peaks of ZnO remain high and visible at the addition of 15%.

To investigate the effectiveness of the composite as antibacterial, the composites were immersed completely in water taken from a traditional well for several days. SEM investigation was conducted to examine the morphology and the presence of any bacteria on the surface of the samples before after the sample immersed into water. Figure 3 shows SEM images of the as prepared samples; (a) 5% Nano ZnO, (b) 10% of ZnO and (c) 15% of ZnO. The pictures show that the surface morphology consists of geopolymers matrix, ZnO particles and other minerals originated from metakaolin. The cracks that become more visible on images (b) and (c) was due to prolonged vacuum during SEM examination. There were no bacteria observed on the surface of the samples.
Figure 3. SEM images of as prepared samples containing different amount of (a) 5%, (b) 10%, (c) 15%

Figure 4 shows the surface of the samples after immersing the samples into water for several days. Sample with 5% nano ZnO appear to form a sodium carbonate crystal which is normally observed on geopolymers [13]. The presence of bacteria can be seen clearly on sample containing 10% and 15% nano ZnO. The type of the bacteria was unknown and need further investigation and consultation. This results indicate that nano ZnO can act as antibacterial agent limiting the growth of bacteria similar to nano Ag.
Figure 4. SEM images of the samples after immersing into water for several days (a) 5%, (b) 10%, (c) 15% of nano ZnO

The mechanical strength of the composite was investigated by means of compressive strength. This measurement is intended to examine the strength of the sample due to the presence of nano ZnO at various concentration. Figure 5 shows magnitude of compressive strength of the samples containing 5%, 10%, and 15% of nano ZnO. The measurements were conducted after samples were stored in open air for 28 days.
Figure 5. The compressive strength of 28 days samples containing different amount of nano ZnO (a) 5 
%, (b) 10 %, (c) 15%.  

Figure 5 shows that magnitude of samples compressive strength fluctuated and decrease as the 
amount of ZnO increase. The addition of 10% of nano ZnO shows the smallest compressive strength 
and increase at the addition of 15%. This discrepancy was probably due to sample preparation or 
measurement error. Time did not permit to repeat the reproduction and measurement of this sample. 
The presence of more ZnO will reduce the binder capability of the matrix and as a result will influence 
the overall strength of the composite. Previous research reported that addition of ZnO into metakaolin 
geopolymers did not improve the compressive strength of the materials [7].

4. Conclusion

A research on the development of geopolymers composite based on metakaolin-nano ZnO as 
antibacterial material has been conducted. The presence of nano ZnO in the structure of geopolymers 
start to appear at the addition of 10%. Immersing the samples into water caused nano ZnO leached out 
from the surface of the sample. SEM micrograph indicated that geopolymers-nano ZnO were able to 
limit the growth of bacteria. The increase amount of ZnO reduces the compressive strength of 
geopolymers.

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