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Morphology of Lightweight Metakaolin-based Geopolymer Polystyrene Composite Paste at Early Ages

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ABSTRACT The integration of polystyrene foams as the lightweight agent in geopolymer is an alternative to produce a lightweight metakaolin-based geopolymer. Meanwhile, the microstructure studies assist to provide better understanding towards the formation of lightweight geopolymer. Hence, the present study examined the morphological characteristics of lightweight metakaolin-based geopolymer polystyrene composite at an early age. The lightweight metakaolin-based geopolymer paste is prepared at ratio 0.7 by using sodium hydroxide as an alkaline activator and the addition of polystyrene foams at ratio of 0.3. The morphology of the lightweight metakaolin-based geopolymer polystyrene paste was analyzed through SEM analysis at 14 days curing ages. The lightweight geopolymer at early ages consisted of the unresponsive metakaolin and porous gel as it gave less dense and compact microstructure. Meanwhile, the analysis showed that the polystyrene foams blended-well in the geopolymer matrix. However, the high temperature during the curing process damages the texture of some of the polystyrene foams. Yet, the utilization of the polystyrene foams as lightweight agents is believed to produce a lightweight metakaolin-based geopolymer.

KEYWORDS: Lightweight geopolymer, metakaolin-based geopolymer, polystyrene foams, geopolymerization, metakaolin.
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Original Article

INTRODUCTION

In recent years, the studies of geopolymer as an alternative in construction materials are getting worldwide attention due to its high properties in compressive strength, fire resistant and chemical resistance. In addition, geopolymer could replace the consumption of cement as the main construction material since the cement manufacturer reduces the greenhouse gases phenomena and sustains the natural sources (Habert *et al.*, 2011). Geopolymer is more known as an environmentally eco-friendly alternative cement compared to Portland Cement due to its sustainability fabrication method and thus reducing carbon dioxide emission that contribute to greenhouse gases (GHG) phenomena and climate change (Habert *et al.*, 2011).

Geopolymer is alumino-silicate based polymers where it contains amorphous form and three-dimensional structures composed of SiO_4 and AlO_4 bonds formed in tetrahedral structure with sharing of oxygen atom that gives various terminologies shown in Figure 1 (Živica *et al.*, 2015; Davidovits, 1994). This inorganic alumina-silicates polymer is generated through the geopolymerization process of mixing alumina-silicate monomers in an alkaline activated solution. The reaction can be happened from the reaction between precursor such as metakaolin (MK) or fly ash-based which rich in aluminosilicate material with the concentrated alkaline activator solution to produce an inorganic polymer binder or adhesive agent (Abdel-Gawwad & Abo-El-Enein, 2016). The most typical raw materials to produce geopolymer composites is metakaolin (MK), fly-ash, kaolin and slag, while the material used to produce the activator solution is composed of sodium hydroxide (NaOH) that is mostly added with sodium carbonate (Na_2CO_3) (Mohajerani *et al.*, 2019; Špaldon *et al.*, 2019; Allahverdi & Mahinroosta, 2019).

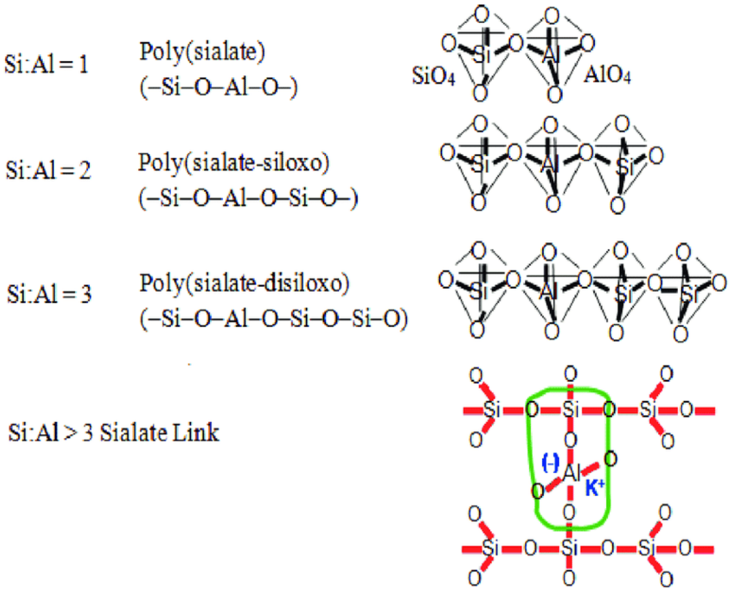

Figure 1. Various terminologies of Si:Al (Davidovits, 1994).

Metakaolin is one of the most effective precursor materials used for geopolymer synthesis that can replace cement material in concrete mixes beside fly-ash, slag and other waste products (Riyap *et al.*, 2019; Kim & Lee, 2020). Metakaolin has a formation of amorphous phases that is produced through a calcination or thermal treatment process of high-purity kaolinite at high temperatures ranging from 700 to 850˚C (Riyap *et al.*, 2019; Kim & Lee, 2020). Previous studies show metakaolin is used as a raw material for geopolymer synthesis due to its highly available material and gives durability strengthening whereas depends on the calcination temperature during the early stages of curing. This durability strengthening that happens at the early stage is due to a combination of the filler effect and increasing cement hydration (Riyap *et al.*, 2019; Kim & Lee, 2020).

Lightweight concrete possesses a low density and frequently used in the construction products in the multi-storey buildings, wide coastal structures, and long bridges because it offers a promising benefit in terms of load bearing elements for smaller cross sections and reductions corresponding to the original size (Riyap *et al.*, 2019). Various lightweight agents have been applied in the products such as foaming agent, sintered fly ash, bloated clay, polystyrene foam and foamed blast furnace slag (Hamad & Hamad, 2014). Polystyrene foam is a light thermoplastic material having a low density, high durability, anti-fungal, low thermal conductor and stable chemical properties such as anti-aging and anti-corrosion (Haibo, 2017; Duan *et al.*, 2017; Mucsi *et al.*,2017). The literature shows the capacity of polystyrene in the development of a low-density concrete (Duan *et al.*, 2017; Mucsi *et al.*, 2017).

Metakaolin-based geopolymer studies are one alternative of cement replacement. Meanwhile, the addition of lightweight agents is seen to enhance the properties of lightweight concrete geopolymer (Riyap *et al.*,2019; Duan *et al.*, 2017). However, studies on lightweight metakaolin-based geopolymer by applying polystyrene foam as lightweight agents still do not give significant numbers especially in the microstructure studies. Therefore, the study on the formation of lightweight metakaolin-based geopolymers polystyrene composite has been conducted. The present study, the morphology of lightweight metakaolin-based geopolymers incorporating polystyrene foams as the lightweight agents at early ages is studied in order to discuss the contribution of polystyrene foams in the microstructure of the lightweight metakaolin-based geopolymer.

METHODOLOGY

Raw Materials

In the study, kaolin is supplied by Sibelco Company Sdn. Bhd. and NaOH pellets (purity ≥ 99.0%, EMSURE®) is used as an alkaline activator of 10 M of concentrated NaOH solution. Polystyrene foams have been used as a lightweight agent in the present study.

Preparation of Metakaolin

Metakaolin is prepared through the thermal treatment of the calcination process of kaolin clay. A weighted metakaolin was calcined through thermal treatment at 800°C with a heating rate of 5°C/min for 2h. The produced metakaolin is then cooled at room temperature and placed in dry condition to avoid the absorption of moisture from surroundings. The mineralogy analysis and FTIR analysis of metakaolin were analysed using Rigaku XRD Analyzer and Perkin Elmer Spectrum One FTIR spectrometer, respectively. The analysis was operated with a step sizes of 0.05^0, scanning rate 0.05^0/min at room temperature, 40 kV and 25 mA in ranging 2^0 to 90^0 of 2θ. In the FTIR analysis, a KBr pellet was prepared at a 1:100 specimen-to-KBr ratio. The FTIR specimen spectrum was obtained in the range 4000 cm^{-1} to 400 cm^{-1}.

Preparation of Lightweight Geopolymer Metakaolin-Based Paste

Lightweight metakaolin-based geopolymer paste was prepared with an alkaline activator-to-metakaolin ratio of 0.7. The binder was mixed until obtaining a homogenous mixture and allowed to rest for an hour. Then, 0.3% polystyrene foam was added to binder as a lightweight agent. The binder was cast in 50 mm × 50 mm × 50 mm cubic molds and cured at 60°C for 24 h. The cubic was stored at room temperature until 14 days of curing ages.

Morphology Analysis of Lightweight Geopolymer Metakaolin-Based.

The morphology of lightweight metakaolin-based geopolymer was analyzed using Hitachi S-3400N SEM. A small geopolymer paste fragments were captured using SEM and operated at 15.0 kV.

RESULT AND DISCUSSION

Mineralogical and FTIR Analysis of Metakaolin

XRD analysis was conducted to analyze the presence of minerals in kaolin and metakaolin. Figure 2(a) and Figure 2(b) shows the diffractograms of kaolin and metakaolin, respectively. In Figure 2(a), the diffractogram of kaolin shows that the kaolinite and quartz are the major phases in this material. The presence of kaolinite can be observed based on the intensity peaks at 2θ= 12.26° and 24.8°. Meanwhile the intensity peak at 2θ= 26.57° indicate the existence of quartz. In Figure 2(b), the diffractogram of metakaolin shows the intensity peaks of quartz at 2θ= 26.57° and 31.3° which indicates that the quartz is a major crystalline phase in this material. In addition, the diffractogram of metakaolin depicts the reduction of kaolinite peaks present compared to the peaks of kaolinite in the diffractogram of kaolin in Figure 2(a). The thermal treatment of kaolin at 800°C activates the breakdown process of the crystal lattice structure through the dehydroxylation of the water molecules that exist in the kaolinite. As a result, it forms an amorphous structure of metakaolin that is rich with an active silica and alumina content. These results are confirmed by the previous studies (Belver *et al.*, 2002; Xu & Deventer, 2002; Merabtene *et al.*, 2019).

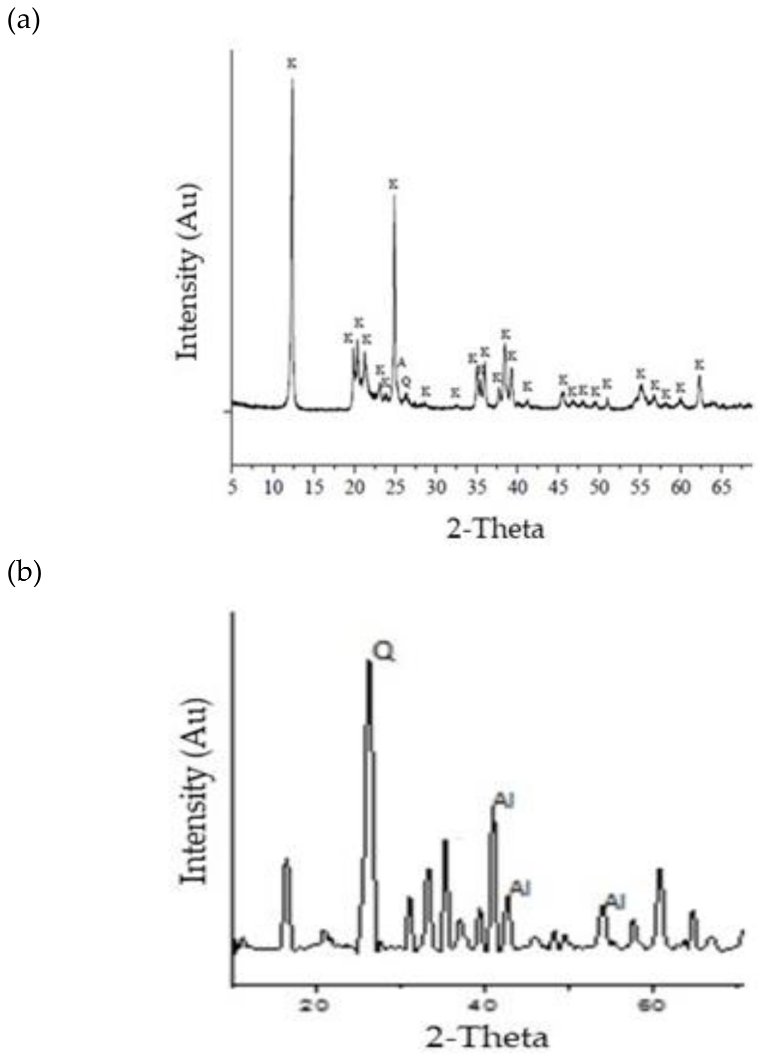

Figure 2. Diffractograms of (a) kaolin and (b) metakaolin obtained at 800°C

The spectrum of kaolin and metakaolin obtained from the FTIR analysis were shown in Figure 3(a) and Figure 3(b), respectively. In Figure 3(a), the appearance of the strong bands ranges from 3683.05 cm^{-1} to 3619.94 cm^{-1} indicates the O-H symmetric stretching vibration as it proves the presence of water molecules. A sharp peak at 992.07 cm^{-1} is observed and the peaks correspond to the Si-O asymmetric stretching vibration, resulting from Si-O-(Al,Si). Meanwhile, the band peaks at 797.39 cm^{-1}, 747.82 cm^{-1} and the strong band 520.31 cm^{-1} correspond to the bending vibrations of Si-O-Al. In addition, the strong peak at 934.01 cm^{-1} indicates the stretching vibrations from Al-O-H. These results indicate the presence of kaolinite and similar with the previous study (Nayak & Singh, 2007).

In Figure 3(b), the appearance of bands at 1023.13 cm^{-1} imply to the Si-O-Si or Si-O-Al asymmetric stretching vibrations. The transformation of kaolin to metakaolin forms an alternating chain between SiO_4 and AlO_4 by sharing an oxygen atom in a tetrahedral coordination (Nayak & Singh, 2007). The bands at 570.40 cm^{-1} and 538.97 cm^{-1} correspond to the bending vibrations of Si-O-Al and the band at 465.23 cm^{-1} indicate the presence of bending vibrations of Si-O-Si. Meanwhile, it is observed that the peak from the stretching vibrations of Al-O-H at 934.01 cm^{-1} did not appear in the spectra of metakaolin. It is suggested that the loss of this peak is attributed to the transformation of kaolin to metakaolin through the calcination process.

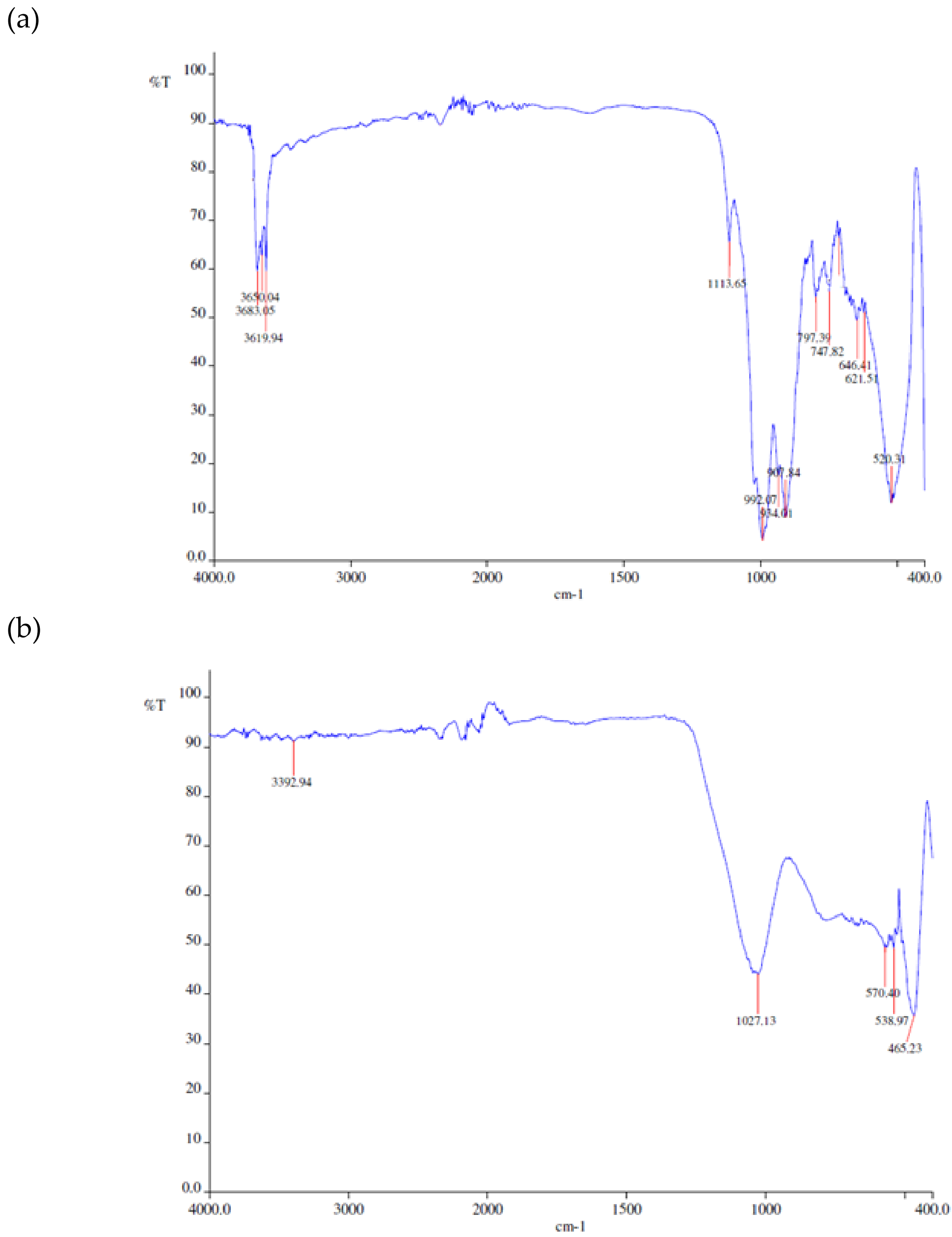

Figure 3. FTIR spectrum for (a) kaolin and (b) metakaolin.

Morphology Characteristics of Lightweight Metakaolin-Based Geopolymer Polystyrene Pastes

Figure 4 (a)-(d) depicts the micrographs of lightweight metakaolin-based geopolymer polystyrene paste at 14 days curing ages. According to Figure 4 (a)-(d), it is observed that the lightweight geopolymer pastes is filled with a homogeneous matrix of unreacted metakaolin, polystyrene foams and polymerization geopolymer products. Meanwhile, it can be seen that the paste of lightweight geopolymer paste on this early age does not give a dense and compact microstructure. The homogenous geopolymer binder is detected in all micrographs and it is a main product of geopolymerization reaction. However, the agglomerate particles of unreacted metakaolin appeared in the geopolymer pastes and can be observed in Figure 4(a) and Figure 4(b). The polystyrene foams also were detected in the pastes as it coated with the geopolymer matrix and some of polystyrene foams are observed with shrink and melt structures. The black holes as it can be detected as the porous gels also appeared on the microstructure of the geopolymer matrix.

Obviously, the dissolution of metakaolin in an alkaline activator produces a homogeneous matrix in paste through the polymerization of aluminosilicate material. The geopolymerization produces an agent binding in the paste which contributes to the main mechanical strength and durability in the geopolymer. Meanwhile, it is suggested that the appearance of agglomerate particles of metakaolin is due to the fact that in the early ages, the metakaolin are likely not completely gone through the geopolymerization process. It also explained the formation of porous gels which represent the capillary gels and it may lead to the cracks of geopolymers paste. It is predicted that these porous gels will reduce at a later age as the geopolymerization of metakaolin continuously occurs in the matrix. This also explains the fact that microstructure of lightweight geopolymer during early age does not provide a dense and compact microstructure. It is confirmed through the SEM analysis that the polystyrene foams are blended-well in the geopolymer matrix. This is an important factor that needs to be considered in the process of integrating polystyrene foam as a lightweight agent so that it can reduce the density of the products. However, it is suggested that the high temperature at 60°C during the curing process gives a possible structure damage to the polystyrene foams as it is confirmed in the results.

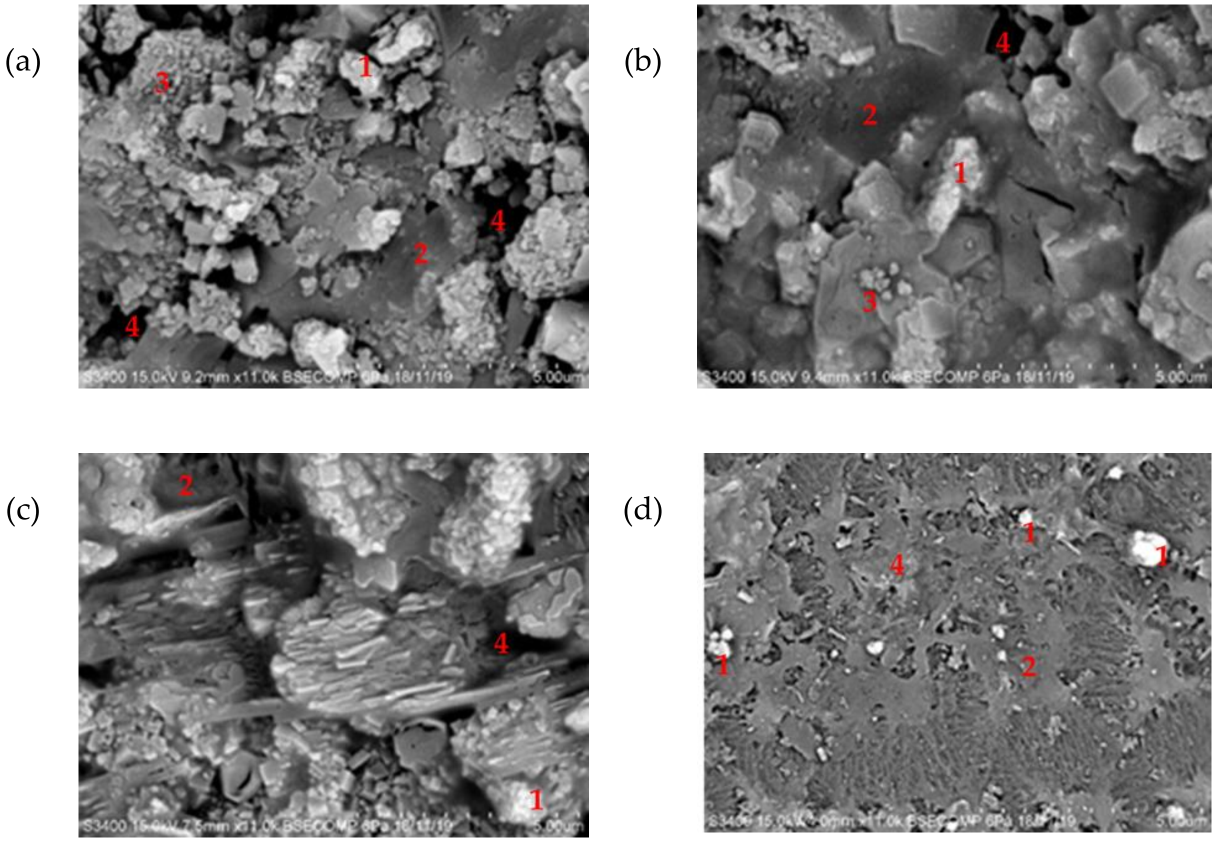

Figure 4. SEM micrographs of lightweight geopolymer metakaolin-based paste at 14 days curing ages. Notation: 1- Polystyrene foam; 2- Geopolymers binder; 3- Unreacted metakaolin; 4- Porous gel

CONCLUSION

Based on the study, it can be concluded that the pastes of lightweight metakaolin-based geopolymer give a less dense and compact microstructure in the early ages which are contributed by the unresponsive metakaolin in the geopolymerization process. However, it is suggested that the microstructure will improve as the geopolymerization process continuously occurs in the geopolymer matrix.

In addition, the results from morphology analysis confirmed that the polystyrene foams are integrated in the geopolymer matrices as the presence of this material will give the lightweight products and reduce the density of the products.

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