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EFFECT OF SOLID-TO-LIQUIDS AND Na₂SiO₃-TO-NaOH RATIO ON METAKAOLIN MEMBRANE GEOPOLYMERS

Geopolymer is synthesized by polycondensation of SiO₄ and AlO₄ aluminosilicate complexes, tetrahedral frames linked with shared silicate oxygen. This paper studies the effect of the solids-to-fluids (S/L) and Na₂SiO₃/NaOH proportions on the preparing of metakaolin inorganic membrane geopolymer. By consolidating a mixture of metakaolin with sodium hydroxide, sodium silicate and foaming agent, the geopolymer membrane was made in required shape about 1 cm and cured at 80°C for 24 hours. After the curing process, the properties of the samples were tested on days 7. Sodium silicate (Na₂SiO₃) and sodium hydroxide (NaOH) solution were utilized as an alkaline activator with a NaOH fixation fixed at 10 M. The geopolymer inorganic membrane tests were set up with various S/L proportions (0.8, 1.0, 1.2 and 1.4) and Na₂SiO₃/NaOH proportions (0.5, 1.0, 1.5, 2.0 and 2.5). Aluminium (Al) powder as a foaming agent was used to create bubbles in porous structure and provide details on the development of membrane geopolymers. This metakaolin membrane, based on the geopolymer, was synthesized by a suspension that depends on the fast cementing mechanism of high-temperature slurries. Porous geopolymeric circles provided a homogeneous composition and quantitative distribution of pores. The water absorption, density, impact toughness testing and microstructure analyses were studied. However, considering the promising results, an adjustment in the mix design of the metakaolin inorganic membrane geopolymer mixtures could increase their mechanical properties without negatively affecting the mechanical properties and porosity, making these sustainable materials a suitable alternative to traditional porous cement concrete.

Keywords: Geopolymer; metakaolin membrane; porous geopolymer; solid-to-liquid (S/L) ratio and Na₂SiO₃/NaOH ratio

1. Introduction

Geopolymer are created by geopolymerization process. Geopolymerization is a green innovative technique for the blend of inorganic polymer material utilizing high silica and alumina content as raw material [1,2]. The case of high silica and alumina content are kaolin, metakaolin, fly ash, dolomite, expanded clay, natural zeolite and slag [3-6]. The first identification proof of geopolymeric materials as an option in contrast to the customary Portland concrete and solid composites in 1979 was proposed by Davidovits [7].

Essentially, the geopolymeric membrane item is from geosynthesis response. It is another innovation of self-supported geopolymeric membrane through geosynthesis response that synthetically integrates minerals which include normally happening aluminosilicate sources to shape 3D tetrahedral

organization [8]. In the alkali solution, hydroxyl ions respond with aluminium and silicon (Si) to isolate aluminate and silicate. From that point onward, the two substances go through a dehydration condensation reaction, whereby a monomer comprising the geopolymer is created. Gelation happens by holding of these monomers, and polycondensation response is repeated to shape geopolymer by curing [9].

Due to strong reactivity, metakaolin is commonly used in geopolymer research. The type of metakaolin is the dehydroxylation of closely bound hydroxyl alloy particles, leading to layered structure and improved mechanical properties of geopolymers [10]. At the stage where kaolin is thermally dehydroxylated in the 500-900°C temperature range, insoluble media dissolves, which makes metakaolin adequate and is a choices for geopolymer production and alternative applications with strong mechanical, physical, chemical and thermal properties [11]. The geopolymer

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characteristics are strongly subject to synthesis parameters such as raw materials, mixing proportions and restore conditions. This work is based on the mixing ratio of the solid to-liquid (S/L) and $\text{Na}_2\text{SiO}_3/\text{NaOH}$ (SS/SH), which are critical in the construction of the geopolymer mechanical quality [11,12]. Yaseri et al. [13] have demonstrated a low viscosity of high S/L geopolymers and the other way in which the disintegration of raw materials is increased.

Ye et al. [14] expressed that a reduction in S/L proportion would expand the smallness of the material just as the mechanical strength. The geopolymer gel is expected to be able to diffuse through the particles into wider interstitial spaces. The separate alumino-silicate particles are thus bound together as the gel hardens, and the resulting matrix presents good mechanical strength. The impact strength likewise affected by the S/L and $\text{Na}_2\text{SiO}_3/\text{NaOH}$ proportion. The lower the impact value speaks to the great impact strength [15]. This is because that low measure of the alkaline activator is deficient to respond with the aluminosilicate source. The mixture of NaOH and sodium silicate formulations (Na_2SiO_3) induces greater geopolymerization concentrations than hydroxide alone for the alkaline activator solution [16]. Sujitra et al., [17] have also shown that different source materials of aluminum-silicate mineral were used for producing geopolymers and for the geopolymerisation process, extra silica (Si) was needed. For disintegration processes of aluminosilicate sources, soluble base hydroxide is needed while Na_2SiO_3 solution is used as a binder [18]. However, excess sodium silicate prolonged the phase of geopolymerisation due to Al-Si precipitation, preventing the interaction of the respondent material with the actuating structure, reducing the activator content [19]. The ideal $\text{Na}_2\text{SiO}_3/\text{NaOH}$ amount to manufacture high-quality geopolymers has been proposed by the researchers in the range 0.67-1.00 [20].

In Stockholm, Sweden, circulated air was first pumped by cement or aerated concrete in the mid-1900s and allocated light-weight or permeable concrete, under which the air was enclosed in a concrete framework. The substance of geopolymer foam displayed a sufficient mechanical property [21]. Hydrogen peroxide is the most well-known foaming agent which contains concrete and water in an autoclave oven in the blend of Portland [22]. Permeable or porous of geopolymer is an elective material for the aerated concrete that needs high vitality in the assembly [23]. Metakaolin, for example, is used mainly as pozzolanic content of porous geopolymers locally available. Elements of permeable geopolymer, for example lime and concrete not containing the combination of geopolymer is special with respect to those of aerated cement. In addition, autoclave method is not necessary for this period as it can be set very well at surrounding temperature and pressure. Therefore, high energy is not required for the processing of the permeable geopolymer, compared with the air circulated by cement, and less ozone-damaging substances transmit to nature [24].

This research accordingly focused around the synthesis of the membrane geopolymer from metakaolin blending in with alkaline activators at different contents the solids-to-liquids

(S/L) and $\text{Na}_2\text{SiO}_3/\text{NaOH}$ (SS/SH) proportions. Those effect on the arrangement of geopolymer based inorganic membrane was investigated for physical and mechanical properties.

2. Experimental method

Materials. In this investigation, Metakaolin, MK was utilized as the precursor, and sodium silicate with sodium hydroxide solutions was utilized as activators. High purity MK was bought from Perak, with a $\text{SiO}_2/\text{Al}_2\text{O}_3$ proportion of 1.6. The chemical composition of metakaolin dictated by X-ray fluorescence (XRF) spectrometer appears in TABLE 2. The activating solutions which are sodium hydroxide arrangement (NaOH) with the concentration of 10 molars arranged from 400 g of NaOH pieces dissolved in 1000 ml of distilled water and colorless fluid of sodium silicate (Na_2SiO_3) which breaks up promptly in water were utilized as soluble base activators for geopolymer base. The aluminium powder as pore-shaping specialists were used in this investigation that encourages the development of foam which would be reasonable for the creation of permeable membrane geopolymer. The aluminium powder was a silver-shaded powder with a particular gravity of 2.7. This is regularly used to acquire hydrogen gas created from substance response that will in general produce hole [25].

Preparations of Metakaolin Membrane Geopolymer.

Alkaline activator solution was framed by blending Na_2SiO_3 and NaOH solutions at a proportion of 0.4, 0.6, 0.8, 1.0 and 1.2 by mass of activator solution until a clear solution was acquired. The sodium hydroxide molarity utilized in this examination

TABLE 1

Mixing design proportions

| S/L Ratio | $\text{Na}_2\text{SiO}_3/\text{NaOH}$ Ratio | Mass of Solid [g] | Na_2SiO_3 Solution [g] | NaOH Solution [g] | Al Powder [g] |
|-----------|---|-------------------|--|-------------------|---------------|
| 0.8 | 0.5 | 100 | 41.67 | 83.33 | 0.2 |
| | 1.0 | | 62.50 | 62.50 | |
| | 1.5 | | 75.00 | 50.00 | |
| | 2.0 | | 83.33 | 41.67 | |
| | 2.5 | | 89.29 | 35.71 | |
| 1.0 | 0.5 | 100 | 33.33 | 66.67 | 0.2 |
| | 1.0 | | 50.00 | 50.00 | |
| | 1.5 | | 60.00 | 40.00 | |
| | 2.0 | | 66.67 | 33.33 | |
| | 2.5 | | 71.43 | 28.57 | |
| 1.2 | 0.5 | 100 | 27.77 | 55.56 | 0.2 |
| | 1.0 | | 41.66 | 41.66 | |
| | 1.5 | | 49.98 | 33.33 | |
| | 2.0 | | 55.56 | 27.77 | |
| | 2.5 | | 59.52 | 23.80 | |
| 1.4 | 0.5 | 100 | 23.81 | 47.62 | 0.2 |
| | 1.0 | | 35.72 | 35.72 | |
| | 1.5 | | 42.85 | 28.57 | |
| | 2.0 | | 47.62 | 23.81 | |
| | 2.5 | | 51.02 | 20.40 | |

was 10 M as the maximal strength was obtained at 10M NaOH solution [26,27]. Metakaolin was mixed in with the prepared activator solution and was stirred well for a few moments. Then, 0.2% by mass of metakaolin of hydrogen peroxide added to the geopolymer slurry as foaming agents to increase the porosity of membrane geopolymer. The detail of mixing extents was given in TABLE 1. Metakaolin membrane geopolymer was produced by a suspension and cementing strategy that created a permeable geopolymeric circle [28,29]. The hardening component of geopolymeric circles came about because of the quick cementing of geopolymers paste at a high temperature which is 60°C that additionally was demonstrated in other past work [30]. The geopolymeric circles could hold round its shape in water bath by quick cementing.

3. Test and analysis method

The study of chemical composition by the X-Ray Fluorescence Spectrometer (XRF) was done by XRF machine, Rigaku Supermini 200. 15 g of powdered sample of metakaolin were put into a plastic sample cup with a plastic support film. This assures a flat surface to the X-ray analyzers and the sample to be supported above the X-ray beam. X-ray fluorescence spectrometer (XRF) is an analysis to obtain data or analyze the raw material element. It is to decide the oxides content and part stage of components.

Water absorption is analyzed, which is the core of the membrane geopolymer, as per ASTM C 140 [31]. Water absorption test is utilized to determine water consumed under certain circumstances. The formula in Equation 1 quantifies water absorption.

$$\left[\begin{array}{l} \text{Water absorption} = \\ = \frac{(\text{Wet weight} - \text{Dry weight})}{\text{Dry weight}} \times 100 \end{array} \right] \quad (1)$$

Next, the density test is used to measure the weight of the metakaolin membrane geopolymer in air and water. It is determined together with other tests, water absorption test. The density of round shape membrane geopolymer was tested by utilizing Electronic Densitometer MD-3005. The samples were kept in encompassing temperature for 24 hours after curing process before test was finished. The unit used in this testing was in g/cm³.

The strength of metakaolin membrane geopolymer was dictated by utilizing impact strength test machine directed by the BS EN 13055-1 standards which is metakaolin membrane geopolymer gets an impact from a standardized dropping weight [32]. The materials were then left to cool down in room temperature before testing are directed. The materials were sieved through 12.5 mm IS sifters. The membrane geopolymer that went through the 12.5 mm strainer includes the test material. The hammering cup of the impact testing machine was fixed in position on the base of the machine. The entire test was placed at the point, and compacted by 25 strokes of the rounded tamping rod. The sample

was removed and sieved through a 2.36 mm IS strainer. The total impact value was then acquired as the ratio of the weight of the fine particles passing through a specific IS sieve to the total weight of the sample, expressed as a percentage as showed in Equation 2. The specification value for the strong impact value or strength is <30% according to their standard. If the aggregate impact value (AIV) is greater than 30%, the results obtained must be treated with care.

$$\left[\begin{array}{l} \text{Impact value} = \\ = \frac{\text{Mass of membrane geopolymer}}{\text{Mass of materials passing}} \times 100 \\ \text{the 2.36 mm sieve} \end{array} \right] \quad (2)$$

To evaluate the samples with porosity, metakaolin membrane geopolymer samples were examined using a high-resolution optical microscope at 100× magnification. It involves magnifying the image of an object by passing or reflecting light away from the object, and then inspecting the light through a single or multiple lenses. Images were generated to evaluate the porosity on the surface of the samples.

4. Result and discussion

Chemical Composition Analysis. From TABLE 2, the chemical composition of metakaolin from XRF investigation was shown that metakaolin contains high measure of silicon dioxide (SiO₂) which was 56.84% and aluminium oxide (Al₂O₃) which is 35.60%. The higher the creation of SiO₂ and Al₂O₃, the higher the reactivity as source materials towards geopolymerisation. Next, the metakaolin composition of magnesium oxide (MgO), sodium oxide (Na₂O) and iron (III) oxide (Fe₂O₃) are very small. There was likewise a little hint of potassium oxide (K₂O), titanium dioxide (TiO₂) and calcium oxide (CaO) where it was not so much critical of the fact that it has under 1%.

TABLE 2

Percentage of chemical compositions of metakaolin by XRF

| Chemical | Mass ratio [%] |
|--------------------------------|----------------|
| | Metakaolin |
| SiO ₂ | 56.84 |
| Al ₂ O ₃ | 35.60 |
| Na ₂ O | 2.40 |
| K ₂ O | 1.04 |
| Fe ₂ O ₃ | 1.31 |
| TiO ₂ | 0.78 |
| MgO | 1.79 |
| CaO | 0.24 |

Water Absorption. The metakaolin membrane geopolymer requires an absorption test since it is a component in evaluating geopolymer durability. Figure 1 indicates the result of water absorbed by metakaolin membrane geopolymer. The results showed that the maximum water absorption rate was at 1.0 S/L

and $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio at 14.98%, while the lowest volume of water absorbed to be at 1.4 solid to liquid by metakaolin mass and 2.5 $\text{Na}_2\text{SiO}_3/\text{NaOH}$, which is just 6.34%. There has been an increase in water absorption from 0.8 to 1.0 with an increase in the solid to liquid ratio, which has fallen from 1.2 to 1.4. The more water penetrates through the concrete, the greater the toughness and strength of the concrete are supposed to be. A porous material that interacts with the surrounding environment is proved by researchers who identified concrete [33]. The greatest influence on the quality of mortar and concrete would be the flow of water into a concrete framework. Because of this issue, the water absorption test is performed to assess the water absorption in concrete. Water absorption shows the porosity level of a substance when the percentage of water absorbed under prescribed conditions is measured. Besides the previously mentioned Zhao et al. [34], increasingly interconnected pores, which are advantageous for water movement, are being used in the end product with the addition of foaming agents in geopolymer slurry. However, the increase in water absorption may reduce the intensity of the impact value. Porosity effects also influence water absorption. The pore size also decreases when the porosity decreases [35]. This is because the composition of the samples is denser, stronger and strengthened. The pore size are reduced and the structure is denser. Analysis in the next section will confirm this. Porosity and permeability also decreased hence the durability potentially be improved.

Density. Figure 2 displays the membrane geopolymer densities of various solids to liquids and the $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio. The densities vary from 1.211 g/cm^3 and 2.831 g/cm^3 . At a solid to liquid ratio of 1.4 and 2.5 $\text{Na}_2\text{SiO}_3/\text{NaOH}$ per mass of 2.831 g/cm^3 are the highest densities reported. A solid-to-liquid 1.0 and $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio of 0.5 by a mass of 1.143 g/cm^3 are found to be the lowest density. The density of metakaolin

membrane geopolymers has decreased from a solid-liquid ratio of 0.8 to 1.0 with a density of 1.294 g/cm^3 to 1.238 g/cm^3 as a bigger pore are formed. In the general sense, it is clear that the metakaolin membrane geopolymer density increased after 1.0 solid to liquid ratio with the rise of solid to fluid and $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio. This is because the volume of solid to fluid required by the combination of alkaline activator is often raised as the solid to fluid is raised. The denser structure may be induced by a decrease of water content according to Bai et al. [36], which is liquid in the geopolymer. Moreover, a medium density was seen to have a rising pattern with the increased $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio. The difference between the maximum average and lowest average density was 1.231 g/cm^3 from 0.5 to 2.5 $\text{Na}_2\text{SiO}_3/\text{NaOH}$. It seems that increases in mean density due to $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio were more uniform. The rise in $\text{Na}_2\text{SiO}_3/\text{NaOH}$ was minimal, due to growing ratios. With the $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio rising, the Na_2SiO_3 amount in alkaline activator solutions has been increased. As more sodium silicate were used because it is denser than NaOH, membrane geopolymer obtained higher density. Sodium silicate rises can lead to more Si ion densification [37-39].

Impact Value Test. The impacts of the solids-to-liquid (S/L) and $\text{Na}_2\text{SiO}_3/\text{NaOH}$ proportions to the impact value of metakaolin membrane geopolymer were analyzed. Figure 3 show least mean impact test value of membrane geopolymer when S/L proportion of 1.2 was utilized at 23.52% and at the S/L proportion of 1.4 demonstrated the highest mean impact strength value which was 33.76%. The most reduced impact value demonstrated that it had the most noteworthy impact strength or toughness out of every other proportion. Mean impact value of S/L proportion 0.8 likewise higher at 31.82%. Even though by expanding S/L, the effect impact value test of membrane geopolymer indicated the ideal proportion of 1.2. This is on the grounds that, the work-

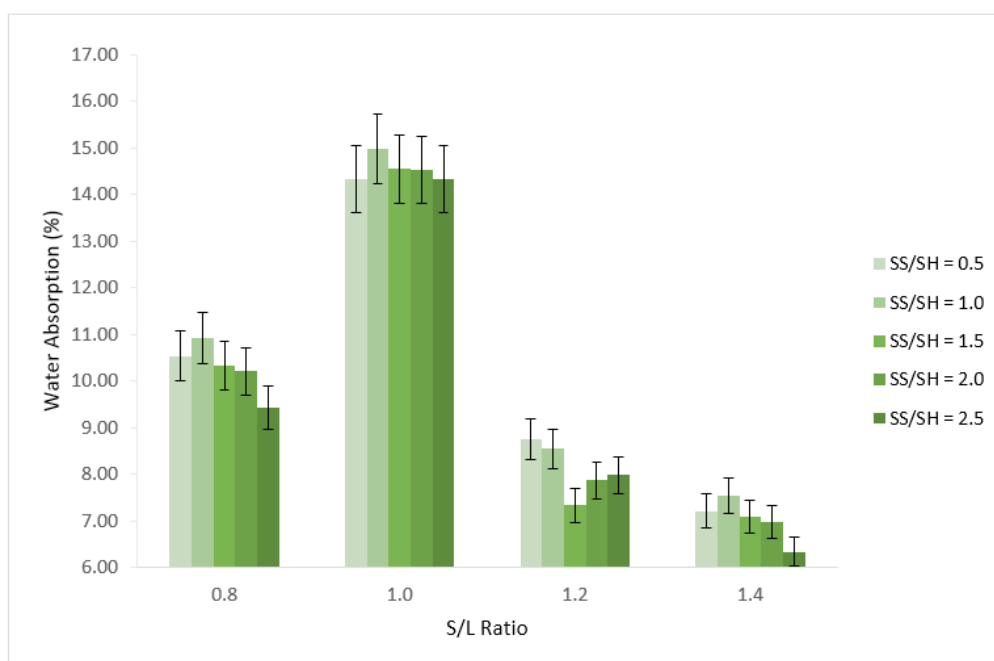


Fig. 1. Water absorption of metakaolin membrane geopolymer with different S/L and $\text{Na}_2\text{SiO}_3/\text{NaOH}$ proportion

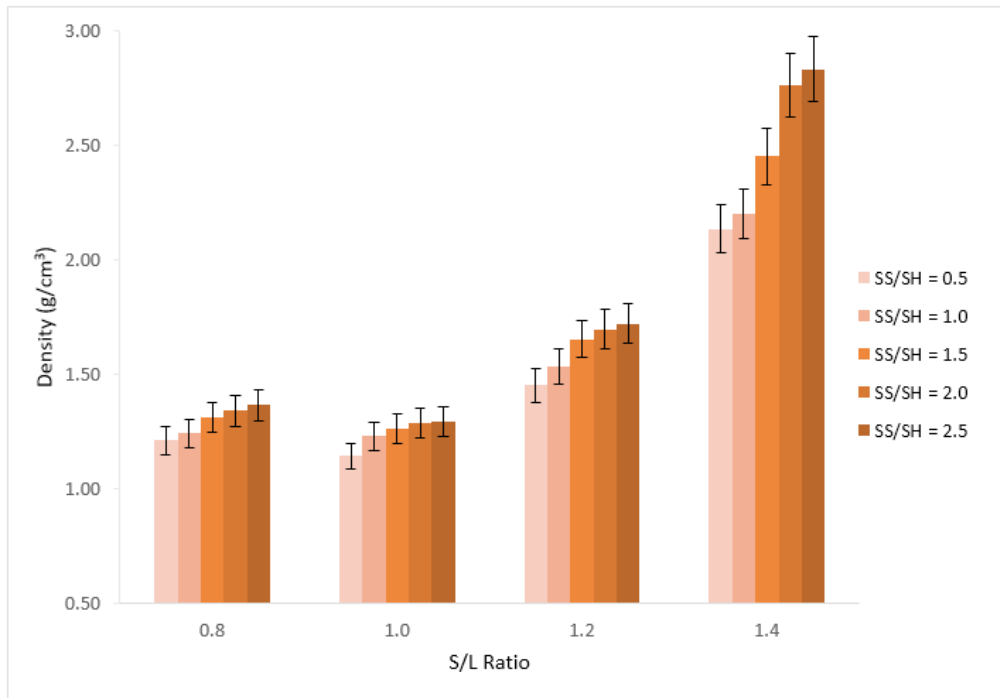


Fig. 2. Density of metakaolin membrane geopolymer with different S/L and $\text{Na}_2\text{SiO}_3/\text{NaOH}$ proportion

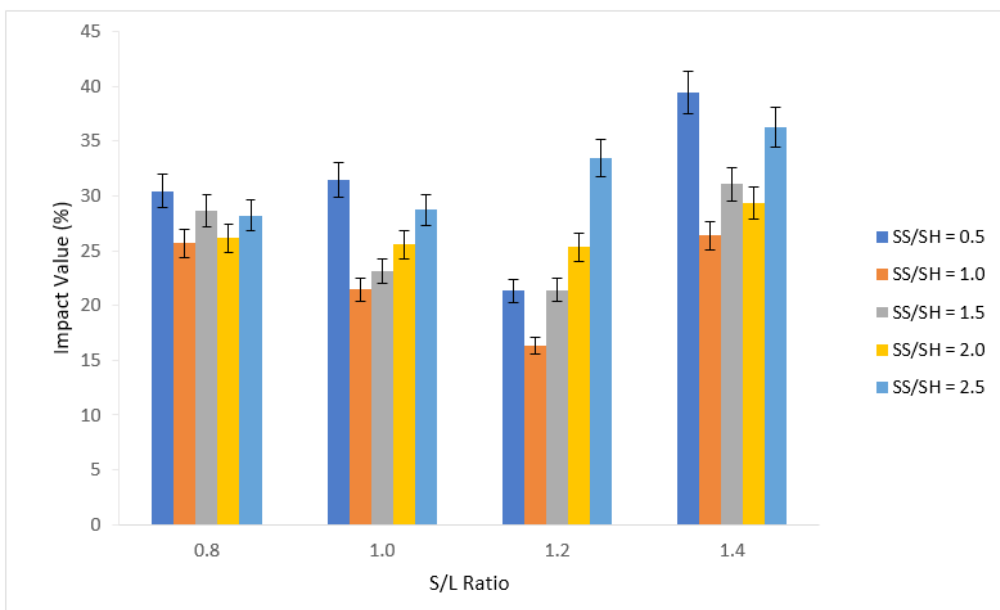


Fig. 3. Impact value of metakaolin membrane geopolymer with different S/L and $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio

ability of the geopolymer with S/L of 0.8 and 1.0 was difficult to palletize into circular shape, it gets stickier and cannot hold round shape after a specific time which demonstrated that the substance of alkaline activator was excessively high. It is likewise hard to give great compaction subsequently lessening the impact value of geopolymer membrane.

Impact value likewise influenced by $\text{Na}_2\text{SiO}_3/\text{NaOH}$ proportion. Figure 3 shows the $\text{Na}_2\text{SiO}_3/\text{NaOH}$ proportion at 1.2 by mass of activator arrangement with the membrane geopolymer mean impact value properties arriving at 22.2%. This was the best worth in general scopes of $\text{Na}_2\text{SiO}_3/\text{NaOH}$ proportions. The

higher the $\text{Na}_2\text{SiO}_3/\text{NaOH}$ proportion, the more the amount of membrane geopolymer is crushed. This implies there is a reduction in geopolymer performance as pavement materials. The vast majority of the outcomes were lower than 30 % impact strength worth proposing that the metakaolin membrane would meet the determination requirements from impact testing standard for use in airfield pavement.

Microstructural Analysis. Computer snap diagram of the surface framework and illustrated in Figure 4. The images in Figure 4 show chosen geopolymer metakaolin membrane whereas

the solid-to-liquid ratios a, b, c, and d are 0.8, 1.0, 1.2 and 1.4 respectively. In this geopolymer slurry, this porous membrane geopolymer had been applied to a fixed volume of Al powder foaming agent. The dark color areas reflect the pores of the sample in all photos, while those which are light colour display the geopolymer matrix. The pores in the sample surface of the microstructure demonstrate clearly the separate solid to liquid effect. There is no major influence on the pores of the sample in $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratios. In either case, in contrast to sodium anions, the potassium anions create more porous structures. These ions effect the workability of the pastes and the final result. Potassium ions are well known to have higher reactivity ratios as a consequence of their higher fundamentality, creating more workable pastes than sodium ions [40]. The alkalinity level of the mixing ratio allows the sample to dramatically enlarge its pores. As previously mentioned, the existence of alkaline ions facilitates a moisture reaction that creates more porous geopolymers. There was a small slurry reaction that resulted in large pores not opening, but still many micropores illustrated the solid-to-liquid ratio at 1.2 (c) and 1.4 (d). The number of pores increases at Figures 4(a) and (b), because of the high alkaline content that can relate similarly to the paste mixture.

5. Conclusion

Effects of the solid to liquid (S/L) and $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratios have been done and discussed on the preparation of the metakaolin inorganic membrane geopolymer. The metakaolin can be used as raw material for the membrane geopolymer

because it contains high reactivity, high silicone and aluminium oxide. Based on the results, when the S/L ratio by mass of metakaolin is 0.8 and 1.0, the sample therefore, shows a greater pore size average, its water absorption is strong and its density is lower, while the S/L 1.2 and 1.4 are less that implies low water absorption and high density. Then, $\text{Na}_2\text{SiO}_3/\text{NaOH}$ proportions also effected to the water absorption and density of membrane geopolymer due to the viscous property of sodium silicate reacting with hydroxide, raw materials and foaming agent that produce the porous sample. To make it clear, image from the metakaolin membrane geopolymer microstructure analysis clearly showed that the sample with growing porosity and pore size obtained the highest absorption of water and low density. Furthermore, the impact strength that has been analyzed for almost this membrane geopolymer obtained a low impact value below 30% with a strong standard characteristic. Overall, the 1.0 solid to liquid mixture by weight and 1.0 $\text{Na}_2\text{SiO}_3/\text{NaOH}$ proportions resulted in the highest water absorption and the lowest density and agreed impact value which are known to be good geopolymer membranes. It may be inferred that porosity inhibits the absorption of liquids. When the porosity reduced, the pores decreased as well. Water consumption declined as well. The sample framework is becoming denser and tougher. The pore size was reduced and the structure are closer to each other. Porous membrane geopolymers are also being formed with high water absorption and lower density that are distinguished by a wide surface and tunable pores that can be used for different media as cost-effective and low energy consumption substitutions since they can be used as an adsorbent, catalyst, sensor, filter and heat insulator.

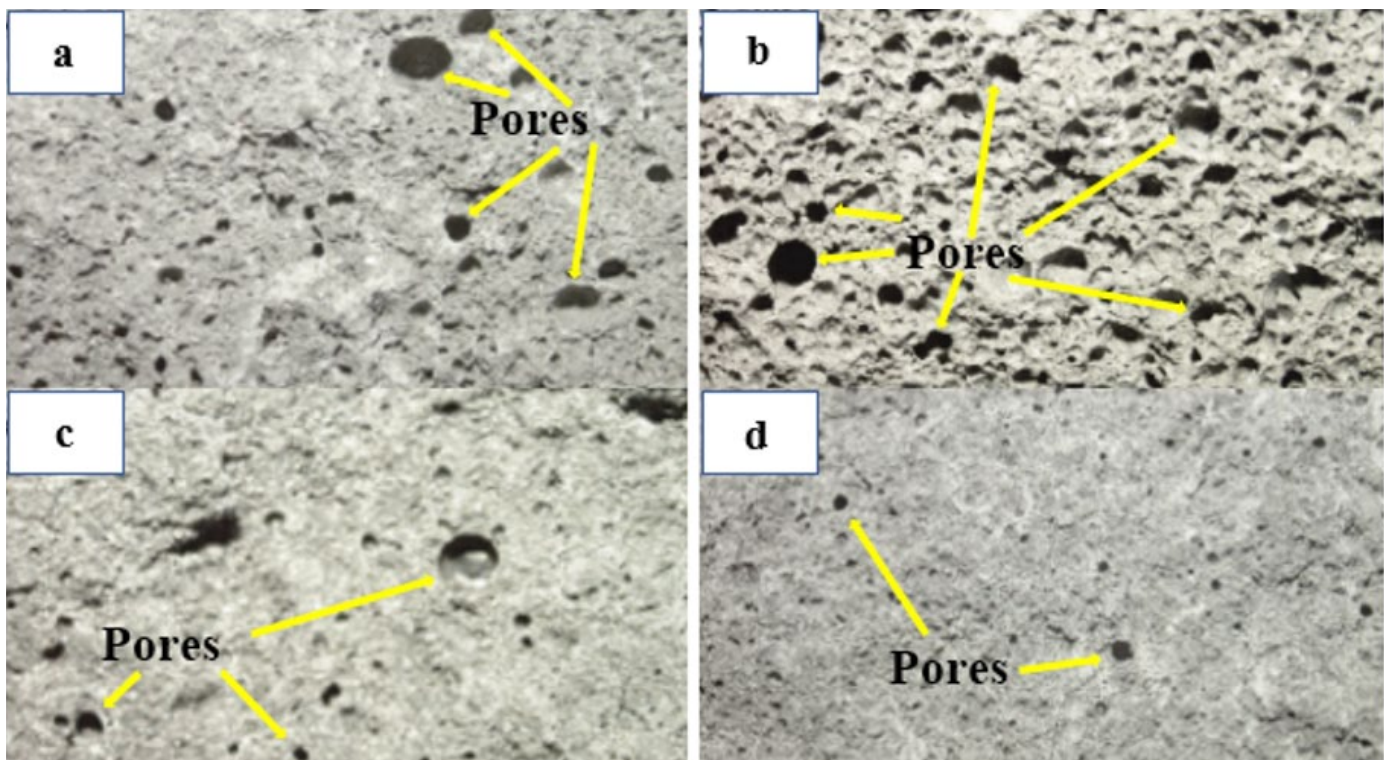


Fig. 4. Microstructure of sample: (a) = 0.8 S/L, (b) = 1.0 S/L, (c) = 1.2 S/L and (d) = 1.4 S/L

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