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THERMAL INSULATION AND MECHANICAL PROPERTIES IN THE PRESENCE OF GLAS BUBBLE IN FLY ASH GEOPOLYMER PASTE

The density, compressive strength, and thermal insulation properties of fly ash geopolymer paste are reported. Novel insulation material of glass bubble was used as a replacement of fly ash binder to significantly enhance the mechanical and thermal properties compared to the geopolymer paste. The results showed that the density and compressive strength of 50% glass bubble was 1.45 g/cm³ and 42.5 MPa, respectively, meeting the standard requirement for structural concrete. Meanwhile, the compatibility of 50% glass bubbles tested showed that the thermal conductivity (0.898 W/mK), specific heat (2.141 MJ/m³K), and thermal diffusivity (0.572 mm²/s) in meeting the same requirement. The improvement of thermal insulation properties revealed the potential use of glass bubbles as an insulation material in construction material.

Keywords: Glass bubble; Thermal insulation; Geopolymers; Fly ash

1. Introduction

Glass bubble is one of newly-explored materials challenges used for numerous applications such as epoxy-matrix composite, filler in cement composite, additive, aggregate and cement replacement [1-8]. This material has been of many interests in many researches due to its unique properties. The material has spherical hollow shape of glass bubble with high crush strength, low density, and easy to work with. Apart from that, the material is also chemically stable, making it become excellent resistance against water and oil. On top of that, its excellent thermal insulation properties have made the glass bubble to be very useful for many industrial sectors including construction industry [9]. The characterisation of glass bubble with hollow spherical shape, low density (125 kg/m³) and low thermal conductivity (0.044 W/mK) provides advantages to geopolymer concrete as new insulation materials. Most of the application of glass bubble as insulating materials in their applications has been reported that the replacement was between 0 to 60%. Table 1 shows the previous reports on glass bubble in several applications and the percentage used along with its measured performance in

concrete applications. Based on the utilization of glass bubble as insulation materials in the literatures, there were no specific guidelines for the best percentage replacement of glass bubble in geopolymer concrete matrix and the finding that provides on how glass bubble performs in geopolymer concrete was not discussed clearly [1-11].

Most of the materials used in concrete production are non-sustainable and contribute to CO₂ emission. As part of the solution, geopolymer materials have emerged as novel engineering materials with the potential to form a substantial element of an environmentally sustainable construction including concrete production [12-17]. With the correct mixture design and formulation, geopolymer concrete can provide superior chemical and mechanical properties to Portland cement concrete, and be highly cost effective [16-18]. One of the main element in geopolymer can be fly ash, obtained as byproduct from power stations, has differ substantially in particle size, morphology and composition because of different coal powders and combustion conditions used [16,19-25]. Fly ash is good for early strength, enhances durability making it suitable to be used as main materials in geopolymer concrete fabrication [19,22-29].

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The development of geopolymer concrete by using glass bubble and having its low thermal conductivity at room temperature and short curing time meets all the quality requirement for concrete production. The unique combination of properties of glass bubble as thermal insulation material and geopolymer concrete however has not yet been. Therefore, a study on thermal insulation properties of glass bubble placement in geopolymer concrete is needed in order to close the gap. Thus, by developing geopolymer concrete with glass bubble as thermal insulating materials, the sustainable green concrete can be achieved with improved thermal insulation properties, and at the same time maintains its strength as outlined in the standard. In this work, the compatibility of glass bubble in geopolymer paste has been studied based on compressive strength and thermal insulation properties to analyze how it can stand and how its performance toward strength and thermal insulation properties in geopolymer system.

2. Experimental details

The compatible of glass bubble in geopolymer paste (Table 1) has been studied at room temperature based on its compressive strength (days of 3, 7 and 28) and thermal insulation properties (28 days). The chosen solid to liquid (fly ash to alkaline activator) ratio is 2.0, the sodium silicate (Na_2SiO_3) solution to sodium hydroxide (NaOH) solution ratio is 2.5 and the molarity of NaOH solution used was 12 M based on the optimum performance by previous studies [30-33].

TABLE 1

The mixed proportion of fly ash geopolymer concrete with replacement variations of glass bubble

Glass Bubble (%)	Fly Ash (kg/m^3)	Sodium Hydroxide (kg/m^3)	Sodium Silicate (kg/m^3)
Control	200	28.570	71.430
2.50	195	27.860	69.640
5.00	190	27.140	67.860
7.50	185	26.430	66.070
10.00	180	25.710	64.290
20.00	160	22.860	57.140
30.00	140	20.000	50.000
40.00	120	17.142	28.572
50.00	100	14.285	35.715

2.1. Materials

2.1.1. Fly ash

The fly ash used in this experiment was acquired from the Manjung power station in Lumut, Perak, Malaysia. The Fly ash is equivalent to ASTM, Class F fly ash (the mean size $46.22 \mu\text{m}$ analysed by particle size analysis) used as the base material for manufacturing geopolymer paste and sieved at maximum $100 \mu\text{m}$

particle sizes (to avoid fly ash particle from agglomerated and enhanced the reactivity in geopolymerisation process).

2.1.2. Glass bubble

The glass bubble or hollow glass microsphere is a ready-product consisting of hollow spherical powdered ultra light inorganic non-metallic materials. Glass bubble is typically formed in pure whitish colour, having high a crush-strength, low-density, high heat and sound insulation, corrosion-resistant, fire resistant and non-conducting. The insulated material chosen was micrometer sized glass bubble (3M. Ltd.), having spherical shapes and extremely light-hollow sphere. The properties of glass bubble in this study are described in Table 2.

TABLE 2

Properties of glass bubbles

Properties	Specification
Composition	Soda-lime borosilicate glass
Colour	White
Density	$125 \text{ kg}/\text{m}^3$
Crushed strength	1.72 MPa
Service temperature	$600 \text{ }^\circ\text{C}$
Median Diameter	$65 \mu\text{m}$
Thermal Conductivity	$0.044 \text{ W}/\text{mK}$

2.1.3. Alkaline activator solution

As the important of alkali activator solution in geopolymer system, the optimum ratio of the alkaline solution is also important where it provides a strong alkaline reaction for solid materials. The combination of sodium hydroxide (NaOH) solution with sodium silicate (Na_2SiO_3) is provides optimum performance in geopolymer system solution with $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio at 2.5 and 12 M molarity for NaOH solution. The NaOH used was in pellet form having 97% purity, and the Na_2SiO_3 consisted of 9.4% Na_2O , 30.1% SiO_2 , and 60.5% H_2O . The other characteristics were: specific gravity at $20^\circ\text{C} = 1.4 \text{ kg}/\text{cm}^3$ and viscosity = $0.4 \text{ Pa}\cdot\text{s}$.

2.2. Methods

The mixing process started by mixing Na_2SiO_3 solution with NaOH solution and stirred until the mixture achieved homogeneity for at least 2 minutes which is adequate for both alkaline solutions to homogenous. The alkaline solution was prepared at least 24 hours prior to use with constant ratio of $\text{Na}_2\text{SiO}_3/\text{NaOH}$ of 2.5. Alkaline solution was added to fly ash for 5 minutes which is commonly applied in producing geopolymer product [34]. Geopolymer paste was then placed in a 50 mm cube mould for compressive-strength testing while for thermal conductivity test was placed in cylinder mould (diameter = 8 cm, height = 2.5 cm).

The samples were later cured at room temperature for 3, 7 and 28 days for compressive strength (ASTM C109) [35], 28 days for density (ASTM C188) [36] and 28 days for thermal insulation (ASTM C168-97) [37].

2.2.1. Compressive strength test

Compressive test is carried out to determine the compressive strength by following ASTM C109 that is using 50 mm cubes specimens. It is important to make sure the face of the specimen that we are applying a straightedge is in good condition before conducting this test. Therefore, the sample is needed to be wiped to a dry surface condition as well as to remove any loose sand grains. This test is started by applying the loads with contact to the specimen. After that, the specimen needs to be carefully placed below the center of the upper bearing block of the compressive machine. Lastly, before recording the result, we need to adjust the data required by compressive machine which are size, weight and specified rate of loading/apply towards the specimens. The total maximum recorded indicated by testing machine and to calculate the compressive strength as follows Equation 1:

$$Fm = \frac{P}{A} \quad (1)$$

Where

- Fm – Compressive strength (MPa),
- P – Total load (N),
- A – Area of loaded surface (mm²).

2.2.2. Density test

The density test of geopolymer paste was conducted on 28 days. The cube geopolymer paste sample of 50 mm was first immersed in water at room temperature for 24 hours and recorded as immersed weight (W_i). The sample is removed from the water and allowed to drain for 1 minute. Then, the geopolymer concrete sample is weighted and recorded as saturated weight (W_s). After that, the geopolymer concrete sample was dried in an oven at a temperature of 110°C for 24 hours then was weighed and recorded as dried weight (W_d). The calculation of the density is explained in Equation 2:

$$\text{Density, } D = \left(\frac{W_d}{W_s - W_i} \right) \times 1000 \quad (2)$$

2.2.3. Thermal insulation

TPS method or thermal constants analyser consists of a variety of transient plane source probes connected to a computerised control unit measure which response as a signal and is sent out to create heat in the sample. These method are distinguished mainly

by the short time (100-120 s) with measurement temperature is –100 to 1000°C required to obtain wide range of thermal conductivity with the needed results (0.016 to 6 W/mK) thus suitable for huge range of different materials. The TPS sensor is placed between two halves of the paste cube sample material. After an equilibration time of at least 45 min in a laboratory nominally maintained at 23°C, measurements were obtained with a power of 0.3 W applied for a measurement time of 10 s.

3. Results and discussion

The volume fraction of glass bubble that had been tested to replace fly ash geopolymer paste was set in the ranges of 0, 2.5, 5.0, 7.5, 10, 20, 30, 40 and 50%. The measured result of compressive strength, density and thermal insulation of geopolymer paste was to determine the suitability and applicable of glass bubble in geopolymer system.

3.1. Density

The measured density of geopolymer paste with various percentage replacement of glass bubble is shown in Figure 1. The figure shows the average density from random sample in the range of 1.450 to 2.155 g/cm³. The highest density (2.155 g/cm³) contributed to the control sample (without glass bubble) while, the lowest density (1.450 g/cm³) is at 50% replacement of glass bubble which contributed of 33% from the control sample. The result indicates that there was a significant reduction in density in the incorporation of high volumes of glass bubble into paste.

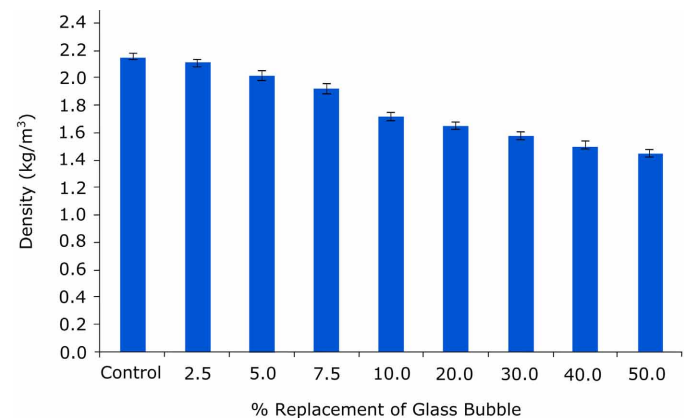


Fig. 1. Measured result of density geopolymer paste with aging days of 28

As one of the main factors in determining compatible of glass bubble in geopolymer paste, density performance also contributes to determine the classification of concrete paste (normal – 2.3 g/cm³, or lightweight 1.0 g/cm³). As can be seen in Figure 1, the density of geopolymer paste decreases as the presence of glass bubbles increases, approaching the lightweight

density. Previous study has obtained densities ranging from 0.5 to 1.7 g/cm³ and thermal conductivities ranging from 0.05 to 0.75 W/mK [38]. As a result, the calculated result density obtained from this geopolymer paste mixture design has a strong tendency to attain low thermal conductivity.

3.2. Compressive strength

The performance of glass bubble in geopolymer paste was viewed based on the compressive strength measured at aging days of 3, 7 and 28 as presented in Figure 2. Increasing the percentage replacement volume fraction of glass bubble has led to a gradual decrease in strength, however the strength was found increases along with aging days. The compressive strength of 50% replacement volume fraction of glass bubble decreases by 10.5%, 18.5% and 29.3% for aging days of 3, 7 and 28 days respectively as compared to the control sample (0%).

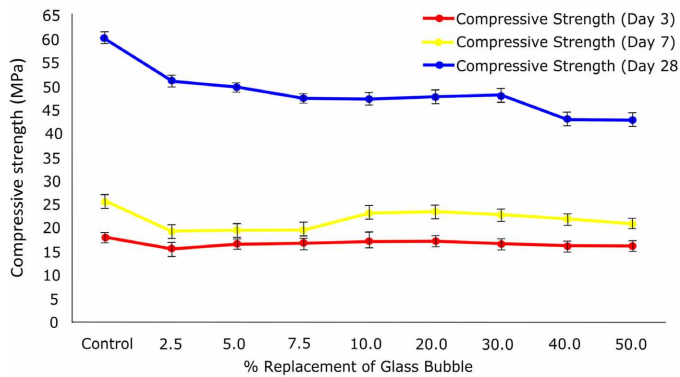


Fig. 2. Measured result of compressive strength geopolymer paste with aging days of 3, 7, and 28

The results of compressive strength have increased with increasing ageing days, indicating that this growth has no durability issues which is in accordance with Khan et al., (2015) [39]. The results of compressive strength with replacement glass bubble shows slightly lower than the control sample (0%) because the structure of geopolymer paste is found to be more compact by the optimum ratio of fly ash based geopolymer paste as reported by Zhang et al., (2015). Meanwhile, the slightly decrease in strength related to the presence of bubbles caused the reduction in the availability of geopolymer matrix formation (the main component contribute or help to good strength) which is in line with findings by Kalaiyarrasi & Partheeban, (2017) [34,40].

3.3. Thermal insulation properties

Thermal insulation properties of geopolymer paste have been studied to understand the behaviour of the glass bubble in geopolymer system.

3.3.1. Thermal conductivity

Figure 3 show thermal conductivity of mixture design geopolymer paste in the presence of glass bubble. The geopolymer paste shows better insulating properties of thermal conductivity depending on the increment of glass bubble amount in the mixture. From the figure, it is observed that additions of glass bubble from 10 to 50% have manifested a percentage reduction of 3.6 to 10% respectively as compared to the control sample (without replacing glass bubble).

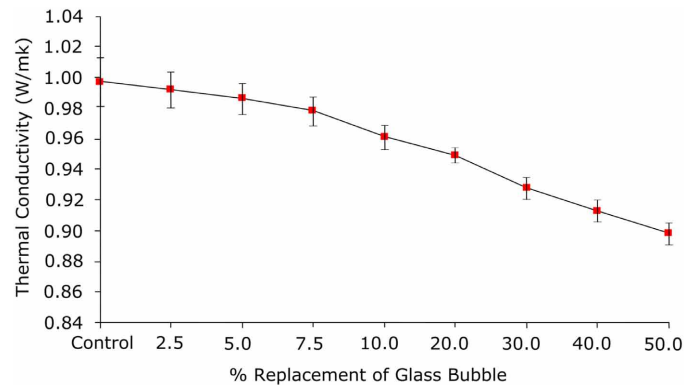


Fig. 3. Measured result of thermal conductivity of trial mix design geopolymer paste

Thermal conductivity values drop, which is understandable given that glass bubbles have a low intrinsic thermal conductivity value (0.044 W/mK) as compared to geopolymer paste since gases are more conductive than solids and liquids. Besides, the presence of glass bubble has enhanced the thermal conduction, as conduction heat transfer in glass bubble does not occur because the diameter particle size of glass bubbles is less than 4 mm, it is reasonable to assume that natural convection of glass bubbles can be ignored when the diameter of the bubble is small [41,42].

3.3.2. Thermal diffusivity

Regarding Figure 4, the figure has shows the measured result of thermal diffusivity in order to perform better understanding on the behaviour of the glass bubble in geopolymer system. With the addition of volume glass bubble into geopolymer paste has influenced the reduction thermal diffusivity value up to 34.5% for 50% replacement as compare to control sample (without replacement glass bubble).

Thermal diffusivity was also investigated in order to create a well-insulated building. The rate at which temperature variations at a material's surface affect the temperature within the material is measured by thermal diffusivity [43]. The lower value of thermal diffusivity obtained with replacement glass bubble is good in delaying the change of temperature inside the building from the outside temperature. In other words, glass bubble has a greater

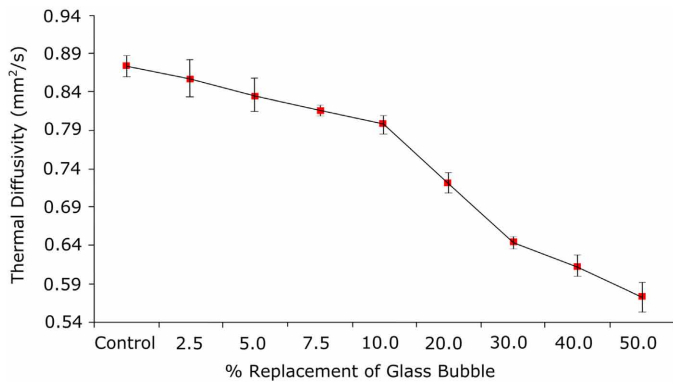


Fig. 4. Measured result of thermal diffusivity geopolymer paste with aging days of 28

thermal resistance, with slower rate of heat transfer. Thus, the thermal insulation material should have low thermal diffusivity to lead the material drop the temperature during a harsh weather and minimizing the heat transfer [44].

3.3.3. Specific Heat

The specific heat of geopolymer paste with increasing percentage of glass bubble is shown in Figure 5. It can be seen from the figure that geopolymer paste with the highest value of specific heat has been dominated by samples with the glass bubble content. The results prove that the specific heat values are in the range of 1.893 MJ/m³K to 2.141 MJ/m³K for 0% (control sample) to 50% replacement percentage volume fraction of glass bubble.

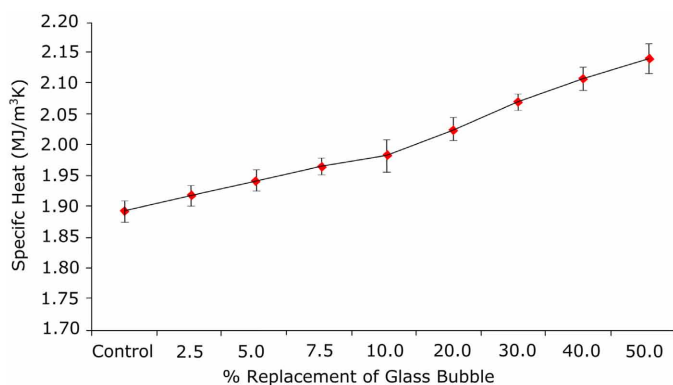


Fig. 5. Measured result of specific heat geopolymer paste with aging days of 28

As the glass bubble replacement percentage rises, the specific heat of geopolymer paste gradually increases due to an increase in the formation of void air of glass bubble leading to a higher specific heat storage capacity. In addition, fly ash as raw material also has little influenced on the measured specific heat [42] if compare to normal concrete only achieved specific heat at 1.0 MJ/m³K with same density. One of the thermal properties

that can be linked to the potential of heat storage capability is specific heat. Since it takes longer for a good insulator to absorb more heat until it heats up (due to extreme weather), it has higher specific heat efficiency [41,42].

4. Conclusions

The compatible of glass bubble in geopolymer paste based on density, compressive strength, and thermal insulation is very important before being applied in geopolymer concrete application. The presented results indicated that the density has decreased with replacement glass bubble as it promises better compressive strength properties as compared to the existing commercial insulated concrete. In terms of thermal insulation properties, geopolymer paste with replacement glass bubble has enhanced properties with the compatibility of 50% glass bubbles.

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REFERENCES

- [1] J.Z. Liang, F.H. Li, *Polym. Test.* **25** (4), 527-531 (2006).
- [2] D. Oreshkin, V. Semenov, T. Rozovskaya, *Procedia Eng.* **153**, 638-643 (2016).
- [3] S. Ren et al., *Mater. Sci. Eng. A* **674**, 604-614 (2016).
- [4] S. Shahidan et al., *MATEC Web Conf.* **103**, 1-9 (2017).
- [5] C. Swetha, R. Kumar, *Mater. Des.* **32** (8-9), 4152-4163 (2011).
- [6] C.Y. Ueki Peres, *Mater. Sci. Forum* **820**, 509-514 (2015).
- [7] K.C. Yung, B.L. Zhu, T.M. Yue, C.S. Xie, *Compos. Sci. Technol.* **69** (2), 260-264 (2009).
- [8] N.F. Shahedan, M.M.A.B. Abdullah, N. Mahmed, A. Kusbi-antoro, S. Tammas-Williams, L.Y. Li, I.H. Aziz, P. Vizureanu, J.J. Wysocki, K. Błoch, M. Nabałek, *Materials* **14**, 809 (2021).
- [9] T.S. Yun, Y.J. Jeong, T.-S. Han, K.-S. Youm, *Energy Build.* **61**, 125-132 (2013).
- [10] Q.-Hu. Q. Bing Liu, Hui Wang, *Materials (Basel)* **11**, 133, 1-16 (2018).
- [11] H. Wang, F. Hou, X. Zhao, *American Journal of Materials Science* **4** (1), 1-11 (2015).
- [12] A. Hanif, S. Diao, Z. Lu, T. Fan, Z. Li, *Constr. Build. Mater.* **116**, 422-430 (2016).
- [13] N. Ariffin, M.M.A.B. Abdullah, P. Postawa, S.Z.A. Rahim, M.R.R.M.A. Zainol, R.P. Jaya, A. Śliwa, M.F. Omar, J.J. Wysocki, K. Błoch, M. Nabałek, *Materials* **14**, 814 (2021).

- [14] E. Nazarimofrad, F. Uddin, A. Shaikh, M. Nili, J. Sustain. Cem. **6**, 1-15 (2016).
- [15] A.U. Zaman, J. Clean. Prod. **66**, 407-419 (2014).
- [16] J. Xie, J. Wang, B. Zhang, C. Fang, L. Li, Constr. Build. Mater. **204**, 384-398 (2019).
- [17] D. Van Dao, H.B. Ly, S.H. Trinh, T.T. Le, B.T. Pham, Materials (Basel) **12** (6), 983 (2019).
- [18] D.D.B. Nergis, M.M.A.B. Abdullah, A.V. Sandu, P. Vizureanu, Materials (Basel) **13** (2), 243 (2020).
- [19] G. Roviello et al., Materials (Basel) **9** (6), 461 (2016).
- [20] J. Xie, J. Zhao, J. Wang, C. Wang, P. Huang, C. Fang, Materials (Basel) **12** (8), 1247 (2019).
- [21] J. Xie, J. Wang, R. Rao, C. Wang, C. Fang, Composites Part B: Engineering **164**, 179-190 (2019).
- [22] S. Sasui, G. Kim, J. Nam, T. Koyama, S. Chansomsak, Materials (Basel) **13** (1), 59 (2020).
- [23] X.Y. Zhuang et al., J. Clean. Prod. **125**, 253-267 (2016).
- [24] W. Ten Kuo, M.Y. Liu, C.U. Juang, Materials (Basel) **12** (10), 1697 (2019).
- [25] U.S. Agrawal, S.P. Wanjari, D.N. Naresh, Constr. Build. Mater. **150**, 681-688 (2017).
- [26] P. Prochon, Z. Zhao, L. Courard, T. Piotrowski, F. Michel, A. Garbacz, Materials (Basel) **13** (5), 1033 (2020).
- [27] L. Biondi, M. Perry, C. Vlachakis, Z. Wu, A. Hamilton, J. McAlozum, Materials (Basel) **16** (6), 923 (2019).
- [28] W.W.A. Zailani, M.M.A.B. Abdullah, M.F. Arshad, R.A. Razak, M.F.M. Tahir, R.R.M.A. Zainol, M. Nabialek, A.V. Sandu, J.J. Wyslocki, K. Błoch, Materials **14**, 56 (2021).
- [29] O.H. Li, L. Yun-Ming, H. Cheng-Yong, R. Bayuaji, M.M.A.B. Abdullah, F.K. Loong, T.A. Jin, N.H. Teng, M. Nabiałek, B. Jež, N.Y. Sing, Magnetochemistry **7** (1), 9 (2021).
- [30] J. Temuujin, A. Minjigmaa, W. Rickard, M. Lee, I. Williams, A. van Riessen, Appl. Clay Sci. **46** (3), 265-270 (2009).
- [31] J. Temuujin, A. Minjigmaa, W. Rickard, M. Lee, I. Williams, A. van Riessen, J. Hazard. Mater. **180** (1-3), 748-752 (2010).
- [32] A.M.M. Al Bakri, H. Kamarudin, O.A.K.A. Kareem, C.M. Ruzaidi, A.R. Rafiza, M.N. Norazian, Appl. Mech. Mater. **110-116**, 734-739 (2012).
- [33] A.R. Rafiza, Y. Zarina, A.M. Mustafa Al Bakri, H. Kamarudin, M. Bnhussain, Aci Mater. J. **109** (5), 503-508 (2012).
- [34] Z. Zhang, J.L. Provis, A. Reid, H. Wang, Cem. Concr. Compos. **62**, 97-105 (2015).
- [35] ASTM C39. Standard test method for compressive strength of hydraulic cement mortars, American Society for Testing and Materials, 2018.
- [36] ASTM C188. Standard test method for density of hydraulic cement, American Society for Testing and Materials, 2018.
- [37] ASTM C168-97. Standard terminology relating to thermal insulating materials, American Society for Testing and Materials, 1997.
- [38] M. Łach, K. Korniejenko, J. Mikula, Procedia Eng. **151**, 410-416 (2016).
- [39] M.I. Khan, K. Azizli, S. Sufian, Z. Man, A.A. Siyal, H. Ullah, AIP Conf. Proc. **1669**, 020065 (2015).
- [40] A.R.R. Kalaiyarrasi, P. Partheeban, Int. J. Emerg. Technol. Adv. Eng. **7** (6), 133-135 (2017).
- [41] B. Nagy, S.G. Nehme, D. Szagri, Energy Procedia **78**, 2742-2747 (2015).
- [42] D. Bentz, M. Peltz, A. Durán-Herrera, P. Valdez, C. Juárez, J. Build. Phys. **34** (3), 263-275 (2011).
- [43] Y.A. Cengel, Heat Mass Transfer A Practical Approach 3rd Edition Cengel, The Unites States, McGraw-Hill., vol. Third edit (2006).
- [44] E. Bwayo, S.K. Obwoya, J. Ceram. **2014**, 1-6 (2014).