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Research Article

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Synthesis of Novel Porous Materials by Geopolymerization Method from Diatomaceous Earth, Rice Husk Ash, and Sodium Silicate Solution

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Abstract This study used geopolymerization technology to develop novel materials with high porosity. Geopolymer is one kind of alumino-silicate materials which was coined by Joseph Davidovits in 1970s. Therefore, raw materials used for geopolymerization normally contain high SiO_2 and Al_2O_3 in the chemical compositions such as rice husk ash, coal ash (fly ash, bottom ash), meta-kaoline, blast furnace slag, red mud, and others. Diatomaceous earth (DE) and rice husk ash (RHA) were used as raw materials with high alumino-silicate resources. Both DE and RHA were mixed with sodium silicate solution for 20 minutes to obtain the geopolymer pastes. The pastes were filled in 5-cm cube molds according to ASTM C109/C109M 99, and then cured at room condition for hardening of the geopolymer specimens for 28 days. The geopolymer-based materials were then tested for engineering properties and porosity using mercury intrusion porosimetry (MIP) method. In which, the engineering properties were tested included compressive strength (MPa), volumetric weight (kg/m^3), and water absorption (kg/m^3). Results indicated that the materials had high porosity over 40% and were considered as lightweight materials with volumetric weight from 642 to 736 kg/m^3; compressive strength at 28 days is in the range of 10.15 to 17.43 MPa; and water absorption is under 236.75 kg/m^3.

Keywords Porous Materials, Geopolymer, Diatomaceous Earth, Rice Husk Ash, Engineering Properties

Introduction

Diatomaceous earth (DE) is one kind of light – weight mineral materials formed million years ago by diatom algae groups. DE contains high silica (60 – 97%) and around 20% aluminum oxides in its chemical composition with amorphous structure. All over the world, DE reserves are predicted as a large amount without specific consideration. The United State, China, and other countries have DE reserving sources about 250, 110 and 550 million tons respectively up to 2012 [1-3]. In another report, total DE reserves of the world are near 918.9 million tons estimated before 2006 [2]. DE is mined and used in fields of filtering materials (67%), cement additive (15%), absorbent (11%), and fillers (7%) [3]. Besides, other applications of DE includes in production food,rubber, paint, insulating material, light-weight material in buiding [4-5]. Vietnam has a large of DE reserve without any solution for mining and applications the material in fact.

Rice husk ash (RHA) is produced by burning rice husk at high temperature. RHA has high porosity with over 80% silica (SiO_2) in its chemical composition. Rice husk takes up about 20% weight of paddy and its compositions include 15 – 20% SiO_2, the others are cellulose and lignin [6-8]. Therefore, after burning process, total weight of obtained RHA is near 20% weight of rice husk. RHA characteristics depend on burning conditions such as temperature, holding time at high temperature. In 2017, world paddy production was at 758.8 million tonnes, in where more 90% belong to Asea countries such as China, India, Indonesia, Bangladesh, Vietnam, Burma, Thailand, Philippine. Hence total estimated RHA reserves are over 30 million tonnes every year [9]. In Vietnam, since 2010, total annual paddy production has been over 40 million tonnes discharged 9 million tons of rice husk without useful solution for utilization of the agricultural waste [9].

Geopolymer is a kind of synthetic aluminosilicate material that is potential field in a variety of applications including a high – performance composites, ceramic and as a replacement for Portland cement [10-12]. Geopolymer is also considered as a green product or environmentally friendly material because of its technological advantages. Hence, it is predicted that this new material can be popularly applied in construction material region in the future [10, 13-14]. The original name that, is "soil cements", was changed to "geopolymer" in the 1970s by Josheph Davidovits [10]. Aluminosilicate minerals are mainly composed of aluminium oxide and silicon dioxide, and some other oxides. Geopolymer is capable to set rapidly and attaining high final strength, as well as having high chemical resistance. Actually, many mechanistic aspects of geopolymerisation provided as potential direction to replace for ordinary Portland cement [10, 11]. The chemical nature of the geopolymer must be determined by control of microstructural development and this is very important in determining the mechanical properties of geopolymer. The thermal and chemical stability of the bonds is determined by nanostructure and molecular structure within the gel phase [10, 15-16].

Both DE and RHA are high porous materials and they are alumino silicate resources consistent with geopolymerization reactions. This research presents the utilization of DE and RHA as raw materials to produce a geopolymer-based material. These raw materials constitute the blend of the alkali-activated binder in this study. DE was used as the primary source of reacted alumina and silicate. Rice husk ash was used as the primary source of reacted silica. It is a by-product of burning agri-waste particularly rice husk, with an estimated generation rate of over 30 million metric tonnes per year worldwide [9, 14]. It is highly porous, lightweight material with very good pozzolanic properties which is used to produce cheap insulating refractory materials (e.g., see [17]).

Materials and Methods

In this paper, raw material of DE was mined from Lam Dong province, Viet Nam. After dried at 110^{o}C for 24 hours, DE was ground in 4 hours by a ball miller and then passed sieves of 90µm.*On the other hand, rice husk was from Mekong Delta, Vietnam and burned at 650^{o}C for one hour in the furnace to obtain rice husk ash. The rice husk ash (RHA) was also ground in 30 minutes and sieved to get particle size distribution under 90*µm*. Sodium silicate solution or water glass solution (WGS) was from Bien Hoa Chemical Factory, Dong Nai province, Viet Nam.*

In this study, mixtures of solid powders with rates of DE and RHA were mixed with WGS concentration from 10 to 30% (in weight of liquid powder per solid solution) using a laboratory cement mixer. Water was added to adjust the paste mixtures to appropriately plasticity enough for workability. **Table 1** showed the mix proportions and WGS solution using for doing experiments in this research. The fresh geopolymer paste was molded to a standard cubic size of 50mm and cured at room condition (30^{o}C, 80% humidity) for 28 days. The geopolymer samples were tested for compressive strength (MPa), volumetric weight (kg/m^{3}), and water absorption (kg/m^{3}) to evaluate effects of WGS to engineering properties of the products. Compressive strength (MPa) and volumetric weight (kg/m^{3}) tests were performed for the 50-mm cube specimens according to ASTM C109/C109M [18]. On the other hand, water absorption test specified by ASTM C140 [19] was also performed. Further investigation, the best sample for the highest strength and lowest water absorption was characterized porosity using mercury intrusion porosimetry method (MIP).

Table 1. Mix proportions used in the design of experiments.

Mixture (Sample)	Proportion of solid powders (%wt)		Concentration of WGS (%wt, liquid/solid)
	DE	RHA	
Geo A1	30	60	10
Geo A2	45	45	10
Geo A3	60	30	10
Geo B1	25	55	20
Geo B2	40	40	20
Geo B3	55	25	20

Geo C1	20	50	30
Geo C2	35	35	30
Geo C3	50	20	30

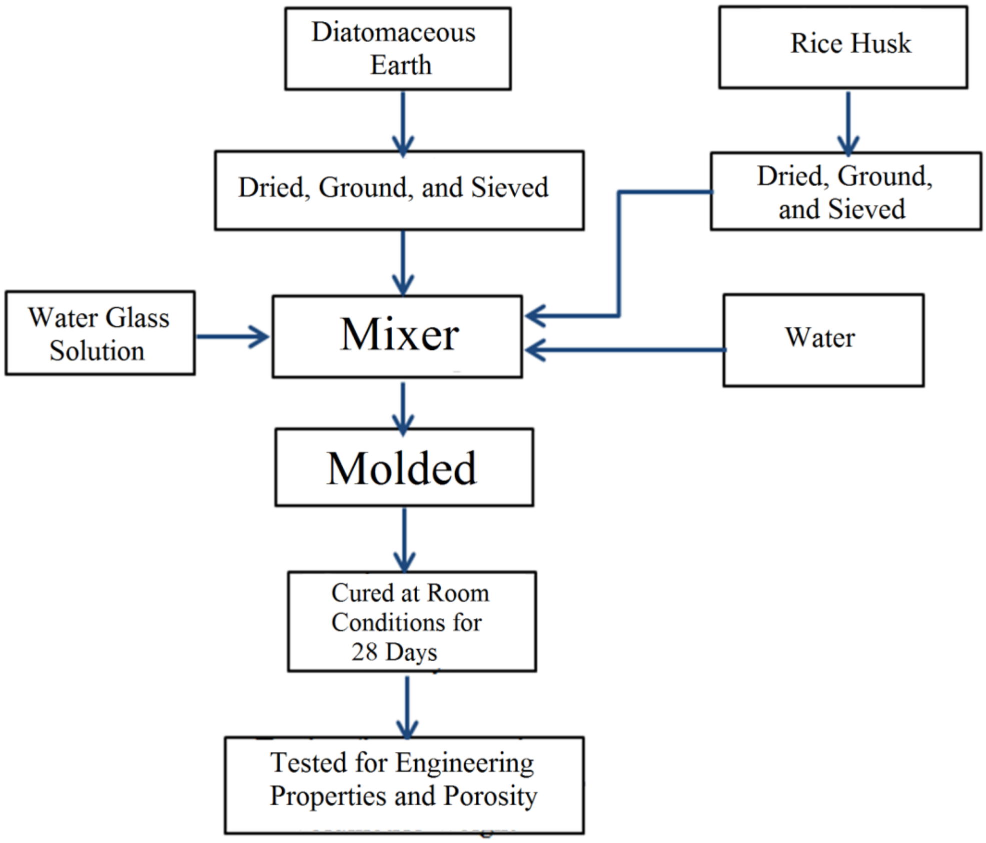

Figure 1: The flow of experimental process

Results and Discussion

Properties of Raw Materials

Table 2 summarizes the chemical composition of these alumino-silicate raw materials. RHA contains high silica with 89.41% of SiO_2 and low loss on ignition (LOI) value at 3.15%. DE has 16.46% of Al_2O_3, 58.16% of SiO_2, 12.07% of Fe_2O_3, LOI value at 8.49% and others in its chemical composition. Figure 2 shows XRD patterns of the materials, both DE and RHA contain amorphous alumina and silica suitable for geopolymerization reactions at high alkaline condition [10-13]. For mineral compositions, DE has quartz, halloysite, nontronite, and kaolinite; RHA contains only cristobalite (SiO_2) in its crystal structure. For the alkaline activator, this study used water glass or sodium silicate solution from Bien Hoa chemical factory which has 32% SiO_2, 12.5% Na_2O and 55% H_2O with a silica modulus of 2.5 was used.

Table 2: Chemical composition (by weight) of DE and RHA

Oxides	**DE**	**RHA**	**WGS**
Al_2O_3	16.46	0.48	-
SiO_2	58.16	89.42	32.00
Fe_2O_3	12.07	1.59	-
Na_2O	2.22	-	12.50
K_2O	0.56	4.83	-
Others	2.04	0.53	-
L.O.I	8.49	3.15	-
Moisture content (%)	5.27	3.18	55.50

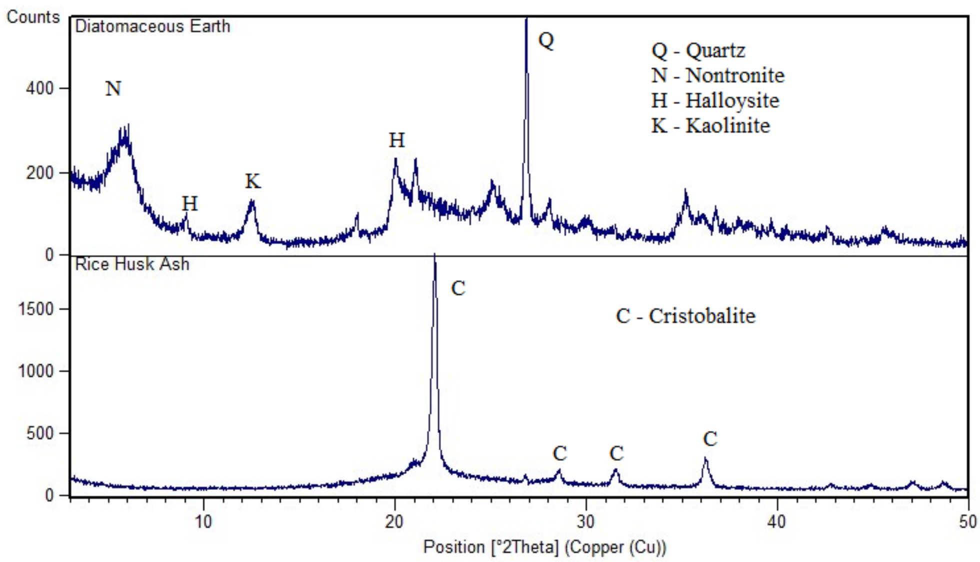

Figure 2: XRD patterns of DE and RHA

Engineering Properties of Geopolymer Products

All geopolymer-based materials have low volumetric weight with the values in range of 642 to 736 kg/m^3 which are less than the prescribed volumetric weight (1680 kg/m^3) for a lightweight concrete brick in ASTM C55-99 and ASTM C90-99a [20-21]. In which, the lowest value of volumetric weight at 642 kg/m^3 belongs to the geopolymer C3 with 30% WGS, 50% DE, and 20% RHA and the highest value of volumetric weight at 736 kg/m3 is of belong to the geopolymer A1 with 10% WGS, 30% DE, and 60% RHA. This is results of using raw materials (DE and RHA) with high porosity and the geopolymerization processes were also produced micro-pores in the geopolymeric structures [10-12].

Table 3: Engineering properties of geopolymer specimen

Samples	Volumetric weight (kg/m^3)	Compressive strength (MPa)	Water absorption (kg/m^3)
Geo A1	736	10.15	236.75
Geo A2	695	12.38	223.16
Geo A3	665	13.17	214.87
Geo B1	698	15.21	229.94
Geo B2	686	16.60	197.53
Geo B3	657	17.43	187.26
Geo C1	703	16.22	186.22
Geo C2	679	16.49	176.29
Geo C3	642	17.18	179.36

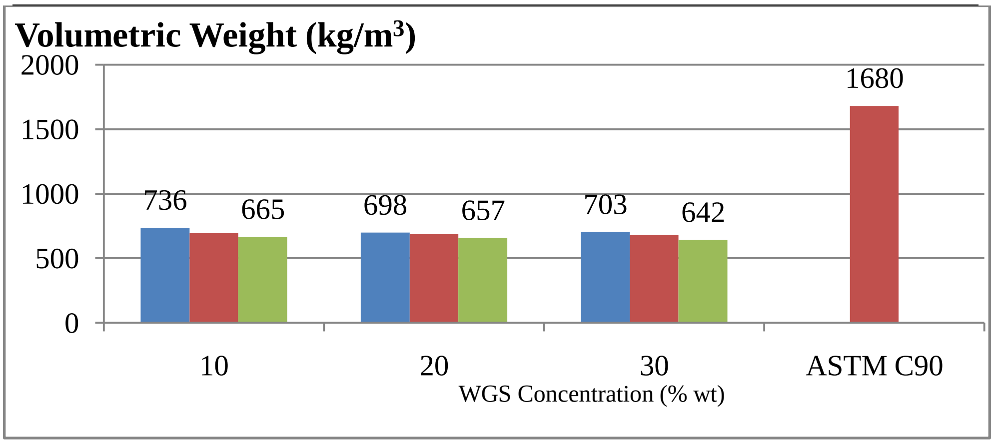

Figure 3: The lower values of volumetric weight compared with ASTM C90 for lightweight concrete brick.

The geopolymer specimens have low water absorption which is in range of 176.29 to 236.75kg/m^3. In which, the lowest water absorption at 176.29 kg/m^3 was of Geo C2 with 30% WGS, 35% DE and 35% RHA whereas the Geo A1 with 10%WGS, 30% DE, and 60% RHA has the highest value at 189.92 kg/m^3 of water absorption. It is noted that the water absorption of the geopolymer-based materials is lower than 288 kg/m^3 which is the prescribed limit according to ASTM C55 or C90 [20-21] requirements for lightweight concrete brick materials.

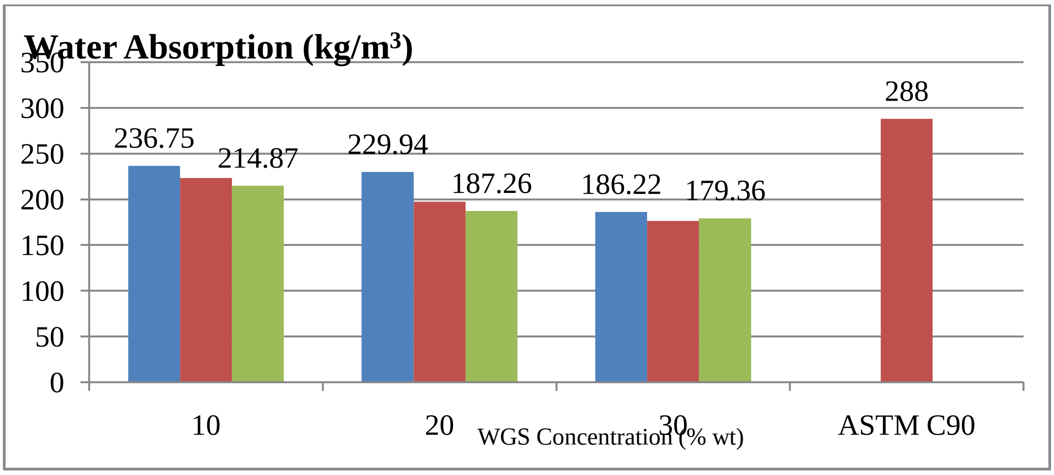

Figure 4: Water absorption of the ash geopolymer compared with lightweight concrete brick in ASTM C90.

The geopolymer-based materials have compressive strength at 28-day in range of 10.15 to 17.43MPa. In which, the specimen of Geo B3 with 20%WGS, 55%DE, and 25%RHA has the highest strength at 17.43 MPa. Most of the geopolymer samples have compressive strength higher than minimum limit of ASTM C55 and C90-99a standards at 11.7 MPa, except for sample of geopolymer A1 (10%WGS, 30%DE, and 60%RHA) with the lowest strength value at 10.15MPa. The results also showed that the best strength of geopolymer-based materials with WGS concentration in range of 20-30%. It is clear that the geopolymerization reactions among activated alumino-silicate resources (in DE and RHA) and alkaline activator (WGS) produced new bondings and networks to make the strength for products.

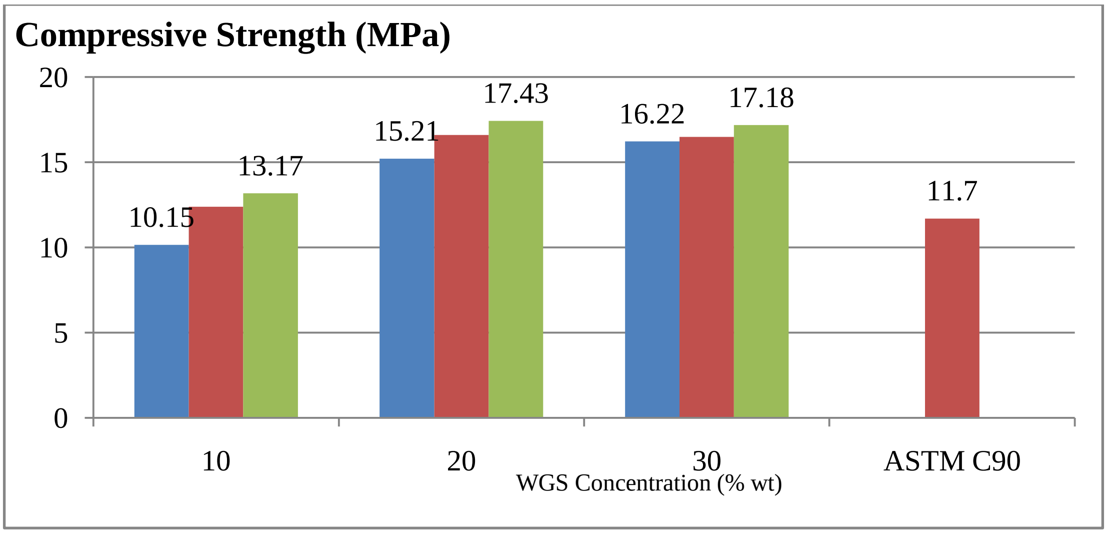

Figure 5: Compressive strength of geopolymer with 10-30% WGS is higher than the lower limits of ASTM C90

The geopolymer sample of Geo B3 had the best engineering properties on low volumetric weight and water absorption (kg/m^3) and the highest compressive strength was carried out for testing porosity using MIP. The MIP result showed that total porosity of geopolymer – based material (Geo B3) is very high at 48.15%. This is the reason why this material has low volumetric weight with bulk density at 0.66 g/cm^3, and the total specific surface area of the porosity is very high at 22.46m^2/g. Note that lightweight concrete ranges from 14 to 34%. The lightweight property of the geopolymer is attributed to its raw material particularly RHA and DE, which are lightweight materials with high porosity.

Table 4: Porous specifications of geopolymer sample B3 tested by MIP

Property	Results
Total cumulative volume (mm^3/g)	343.17
Total specific surface area (m^2/g)	22.46
Total porosity (%)	48.15
Bulk density (g/cm^3)	0.66
Apparent density (g/cm^3)	2.24

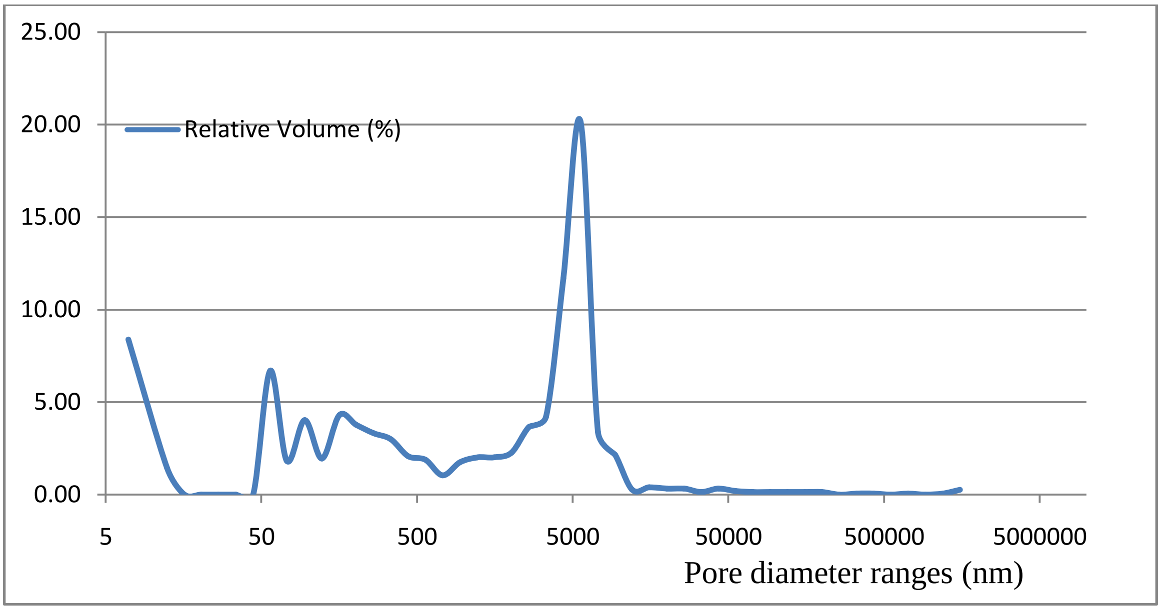

Figure 6: Pore size distribution of geopolymer – based material, sample Geo B3

The pore size distribution of the geopolymer – based material (Sample Geo B3) is in large range of 7nm to 2.0nm as shown in Figure 4. In which there are under 24% pores with diameter from 5nm to 0.1 µm, around 25% pores with diameter from 0.1 µm to 1 µm, around 50% pores with diameter from 1 µm to 10 µm, and under 1% pores with diameter over 10 µm for relative volume. There are several reasons to explain the formation of pores inside the geoplymer. One could be attributed to the pores existed from raw materials (De and RHA). Pores could also be formed after geopolymeric reactions (i.e., void space formed from the evaporation of water or reaction of water with solid particles). Void spaces could also be formed from the mixing and molding process if solid particles have not been compacted well and air bubbles are trapped in the wet geopolymer mixture.

Conclusions

Synthesis of geopolymer-based materials is always interesting topic for researchers. This study introduces a new geopolymer with high porosity produced from a blend of diatomaceous earth and rice husk ash using water glass solution (sodium silicate solution) as an alkaline activator. The porous geopolymer-based materials are good performance with engineering properties responded to requirements of ASTM C55 and C90 for lightweight concrete brick. In which, the porous geopolymer with a solid powder mix of 55% DE and 25% RHA and alkaline-activated with 20% WGS had an average 28-day compressive strength of 17.43 MPa, water absorption of 187.26 kg/m^3, volumetric weight of 657 kg/m^3. The geopolymer-based material also has high porosity at 48.15% with the pore size distribution under 10µm. Hence, the geopolymer-based materials can be potentially used as lightweight material for masonry walls or partitions.

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