Effects of Foaming Agent Contents and Firing Temperatures on Porosity and Density of Granulated Foam Glass Prepared from Waste Glass

ผลของปริมาณสารให้โฟมและอุณหภูมิในการเผาต่อความพรุนตัว และความหนาแน่นของแก้วโฟมแบบเม็ดซึ่งเตรียมจากเศษแก้ว

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Received: Sep. 11, 2019 **Revised:** Nov. 12, 2019 **Accepted:** Dec. 17, 2019

Abstract

The purpose of this research work was to analyze production of granulated foam glass (GFG), a lightweight aggregate used in cement concrete, using mixed waste glass as the main ingredient. The effects of foaming agent content and firing temperature on density and porosity of the GFG were studied. Dolomite mineral and tapioca gel were selected as the foaming agent and the binding agent respectively. Dolomite content was varied from 1-4 wt % while the tapioca gel was fixed at 35 wt %. Firing temperatures used were 750 °C and 800 °C for 10 min. It was found that firing at 750 °C was more suitable than at 800 °C as the latter temperature led to non-uniform porosity due to bloating. When increasing the dolomite content, the bulk density of the GFG decreased while the porosity increased. The best condition obtained from this current research was 3% dolomite and 750 °C firing as the GFG showed a desirable bulk density of 0.54 g/cm^3 and a porosity of 76% with a uniform pore structure. The pore size of this GFG was mostly found to be in the 10 μm range.

Keywords: waste glass, granulated foam glass, density, foaming agent content, firing temperature

1. Introduction

Waste glass has become an urgent environmental problem all over the world due to its landfill spaces occupancy. The waste glass was regarded as " dangerous waste" in Thailand so the elimination of this waste needs budgeting from local municipality.

In practice, glass recycling occurs at a lower percentage than expected due to limitation of the recycling process. In order to solve this problem, the use of waste glass in other applications especially in construction has been proposed by many researchers (Serpa et al., 2013, 489, Corinaldesi et al., 2005, 197, Rashad et al., 2014, 340). Waste glass may be crushed into desired size and added into the concrete recipe directly in order to reduce the amount of fine aggregate used (Islam, et al., 2017, 38). Another approach of the utilisation of waste glass was to use as the aggregate in concrete for a more advanced purpose of thermal efficiency as well as thermal insulation (König et al., 2018, 11143). In the method, waste glass was prepared as "granulated foam". Granulated foamed glass (GFG) has a lifespan equal to that of construction materials, e.g., bricks and concrete (Kaz'mina et al., 2011, 371). Foamed glass is becoming increasingly common as an effective thermal insulation material.

Foam glass is a heterogeneous phase system consisting of the solid and gaseous phases with low density, low thermal conductivity, low vapor impermeability, no water absorption, incombustibility, good chemical resistance so I can be used as lightweight (Chen et al., 2012,264).

The types of common foaming agents such as carbon, calcium carbonate ($CaCO_3$), dolomite and silicon carbide determine the composition of released gases (Souza et al., 2017, 61). The amount of foaming agents used is dependent on the amount of gases released and viscosity of the melting materials. Examples of the foaming agent used are such as eggshell (1 - 30%) (Souza et al., 2017,60) to foam waste glass bottle or 1% $CaCO_3$ to foam waste glass cullet (Stiti et al., 2011, 3384).

The foaming agent must decompose at the desired temperature, making retention of the foamed structure possible. In order to achieve low density and uniform pore structure, it is necessary to avoid collapses through coarsening of individually created bubbles. Coarsening of bubbles occurs to reduce free surface energy. To improve properties especially strength of the GFG, many raw materials such as fly ash, rice husk ash, may be used as axillary materials in preparations (Chen et al., 2012, 263, da Silva Fernandes et al., 2019, 77, Wang et al., 2018, 13681).

Naturally occurring dolomite-a double carbonate of Ca and Mg-CaMg $(CO_3)_2$ is typically sedimentary rock associated with calcite, widely scattered in nature (Gunasekarn and Anmalagan, 2007). The decomposition of dolomite started around 584 K and ended around 1119 K with multi-step reactions (Olszak-Humienik and Jablonski, 2015) so it was suitable to use as the foaming agent for the mixed glass types which sintering could occur at a various temperature range. The amount of dolomite used as the foaming agent in transparent waste glass reported was up to 5% (Pokorny et al., 2011, 174)

In the present study, mixed waste glass from local municipals was used as the main material for the preparation of GFG. Natural dolomite was used as the foaming agent. Tapioca gel, because of its low price, was used as the binder for shape forming. Preparation and characterization of the GFG are addressed in this paper.

2. Objectives

2.1 To study the potential use of mixed waste glass as the raw material for the preparation of granulated foam glass.

2.2 To study the effects of foaming agent content and firing temperature on porosity and density of the GFG.

3. Materials and Methods

3.1 Materials Characterisation and Preparation

The mixed waste glass was collected from Sansai municipality, Majan District, Chiang Rai. Dangerous glass such as CRT tubes or fluorescent lamp envelopes was separated, so mainly soda-lime glass was used as main starting material. The collected glass was first clean and handed crush using a solid bar. After that coarse grinding of the crushed glass was done using a Hammer milling, followed by ball milling for different lengths of time to obtain the desired particle size. The glass powder preparation steps are schematically displayed in Figure 1. The particle size of the milled glass was analyzed using sieve analysis method, the result is graphically shown in Figure 2 (a) and an example of glass powder particles taken by an optical microscope is also shown (b). From the literature, GFGs with a density of <150 kg m^{-3} were obtained when the particle size is ≤33 μm (D50) (König et al. 2016, 190). The milling time selected was thus 8 hrs.

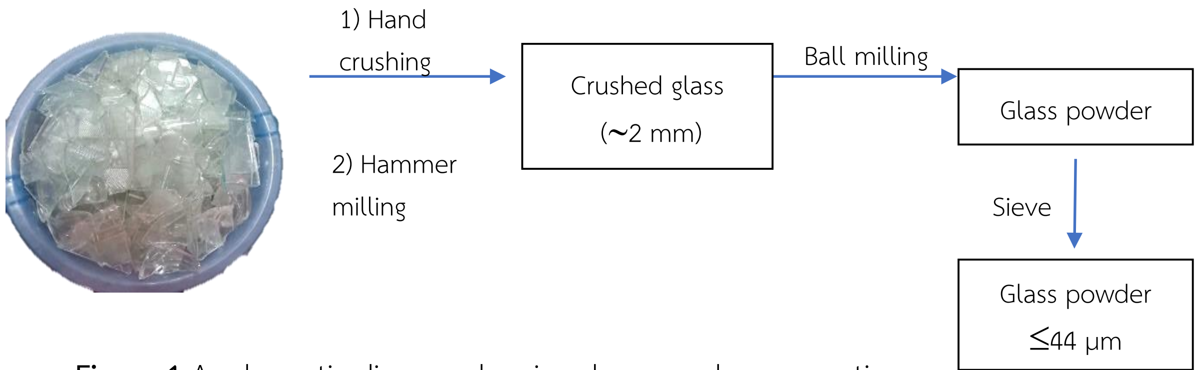
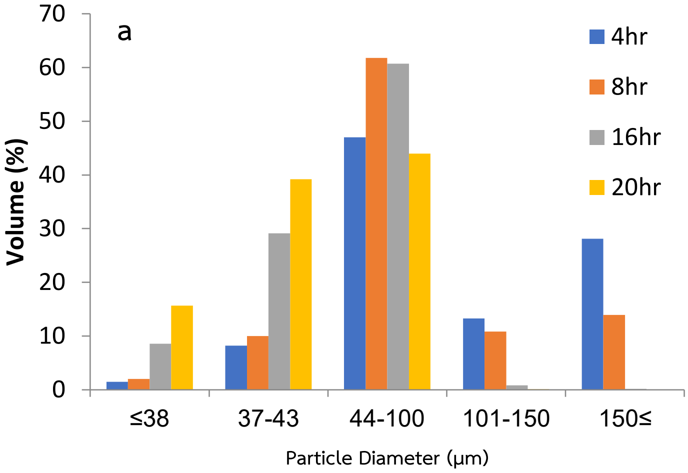


Figure 1 A schematic diagram showing glass powder preparation



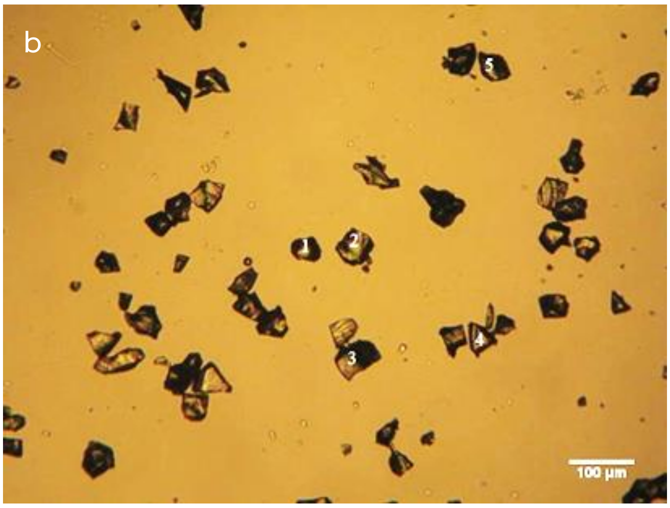


Figure 2 (a) Sieve analysis result of the glass as a function of ball milling time, (b) OM picture of the glass particles passed through the sieve #325 mesh-sieve after ball milling for 8 hrs.

Dolomite clay which decomposes and gives out CO_2 gas was chosen as the foaming agent. Chemical compositions (wt. %) of the waste glass and dolomite clay were analyzed using X-ray fluorescence (XRF) and are shown in Table 1. X-ray diffraction of the dolomite clay in Figure 3 indicated the existence of dolomite (JCPDS:83-150) with some calcite mineral (JCPDS:5-586). Decomposition of dolomite is shown in Equation 1 (Olszak-Humienik and Jablonski, 2015, 2239):

$$(CaMg(CO_3)_2) \longrightarrow CaO(s) + MaO(s) + 2CO_2(g) \qquad (1)$$

Table 1 XRF chemical compositions (wt.%) of waste glass and dolomite.

Raw material	SiO_2	CaO	Na_2O	MgO	Al_2O_3	K_2O	Fe_2O_3	TiO_2	SO_3	ZrO_2	SrO
Waste glass	72.10	12.35	9.29	3.74	1.23	0.44	0.42	0.11	0.20	0.04	-
Dolomite clay	0.71	80.05	-	19.01	-	-	0.19	-	-	-	0.04

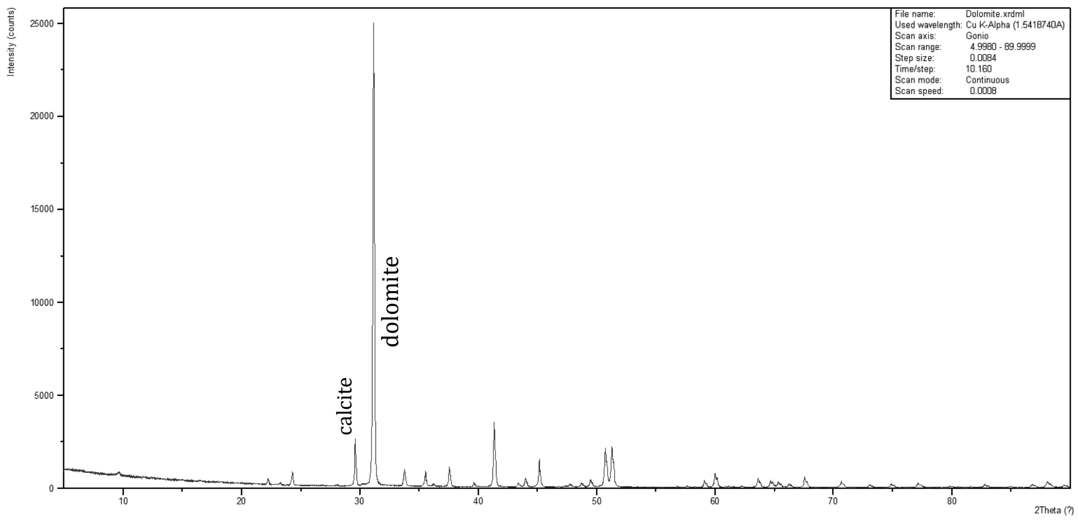


Figure 3 X-ray diffraction analysis of dolomite clay indicating the dominant mineral of dolomite with some calcite coexistence.

3.2 Granulated Foam Glass Preparation and Their Property Testing

In order to select the proper preparation condition, dolomite was first mixed at 1, 2 and 3% to the waste glass to make up the total weight of 100 g. The raw materials were mixed using a mixing machine for 3 min before being compacted using uniaxial dry-pressing at 10 MPa into disks with a diameter of 5 mm. From our previous studies, waste glass could be sintered at around 750 °C (Wattanasiriwech et al., 2019, 012008) while some literature reported the sintering temperature of 800 - 900 °C (Stiti et al., 2011, 3384) (Souza et al., 2017, 61). The samples were thus sintered at 750 and 800 °C with a heating rate of 5 °C/min, 10 min dwell time and a natural cooling.

Granulated foam glass (GFG) was prepared with the aids of tapioca gel as the binding agent to suit the commercial manufacturing process. Glass powder was first mixed with 1 - 4 wt% dolomite using a mixing machine. Tapioca gel was later mixed 35 wt% to the mixture before granulating to spheres of around 5 mm. in diameter. The granulated glass was fired at 750 °C with a heating rate of 5 °C/min and 10 min dwell time.

Bulk density (ρ) and true density (Q) of GFGs were measured based on 10 samples, following ISO 5016:1997 (Shaped insulating refractory products - Determination of bulk density and true porosity). Bulk density was calculated according to Equation 2:

$$\rho = m/v_{GFG} \tag{2}$$

Where: m is mass of each GFG, V_{GFG} is the volume of the GFG determined using water displacement when GFG was added to the water. V_{GFG} is the volume difference between the immersed water and original water measure in the 10 mL cylinder.

True density (Q) of the GFGs was measured using a Pycnometer with a capacity of 25 ml. The true density was calculated according to Equation 3:

$$Q = \frac{m_1}{m_3+m_1-m_2} \times Q_{liq} \tag{3}$$

Where: Q is true density, Q_{lig} is the density of the liquid, m_1 is initial mass of test material, m_2 is mass of the pycnometer filled with a quantity of the test material and with test liquid, m_3 is the mass of the pycnometer filled with the liquid used. True density of the GFGs averaged from 3 samplings was 2.29 g/cm^3.

The porosity of the glass foam was measured following the same standard above. Porosity, π_1 is the ratio of the total volume of the open pores and the closed pores in porous body to its bulk volume. Percent true porosity, π_1, is given by Equation 4 (Chen *et al.*, 2012, 264):

$$\pi_1 = \frac{\rho_t - \rho_b}{\rho_b} \times 100\% \tag{4}$$

where: ρ_t are the true density of the foam glass (g/cm^3), ρ_b are bulk density of the foam glass (g/cm^3)

4. Results and Discussion

3.1 Effects of the dolomite content and firing temperature

Figure 4 shows GFGs prepared with different dolomite clay amounts (denoted as GFG1, 2, 3) and two firing temperatures of 750 °C and 800 °C (denoted as GFGx-750 and GFGx-800 respectively where x represents the amount of dolomite clay addition). The GFG was compacted without any binder. The result suggested that firing at 750 °C was more suitable than at 800 °C as the porosity was more uniform and bloating was not yet observed. At 750 °C, foaming was more progressive with increasing the dolomite contents, as it was suggested by the size change and also by the thinner crust thickness. Glass expansion was caused by the release of decomposition gas from the pore-forming agents when it showed suitable viscosity (da Silva Fernandes, 2019, 80)

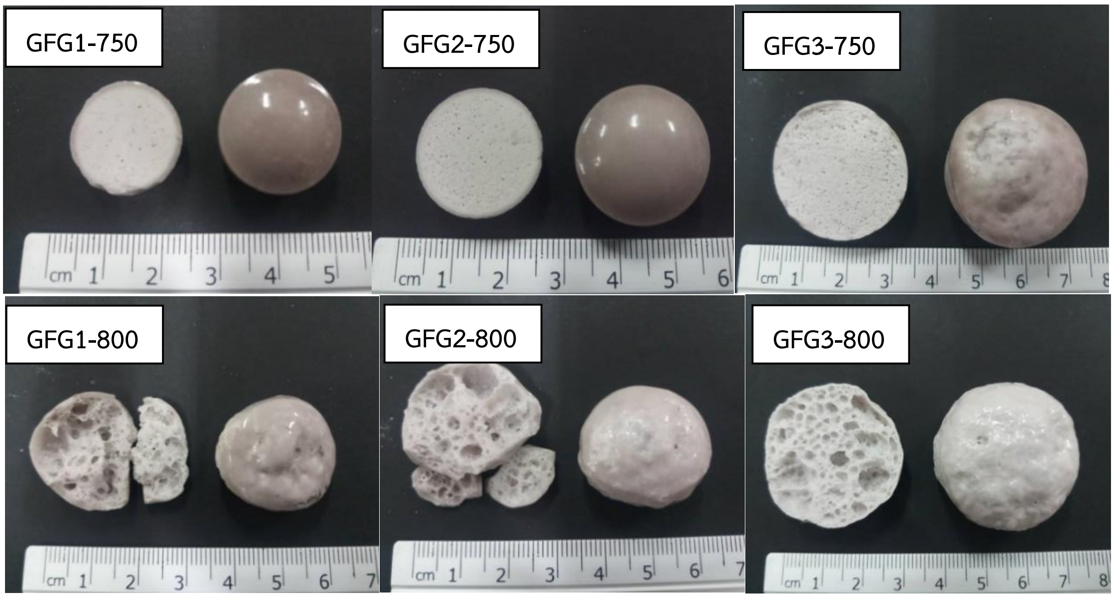


Figure 4 Effects of dolomite clay content and firing temperature on foaming characters of the GFG prepared by compaction.

Figure 5 shows GFGs prepared with different dolomite clay amounts (denoted as GFG1, 2, 3, 4) and with 35% tapioca gel. The GFGs were fired at 750 °C At 1% dolomite clay, foaming had only slightly occurred. With Increasing the dolomite clay content to 2 or 3%, foaming became more uniform, but when further increasing the dolomite clay content to 4%, some bloating could be seen.

Bulk density as a function of dolomite content in the GFGs prepared with tapioca gel binder is graphically displayed in Figure 6. The result shows that with increasing dolomite contents from 1 to 2%, bulk density was dramatically reduced from 1.55 g/cm^3 to 0.87 g/cm^3 and continuously decreased to 0.58 g/cm^3 and further to 0.39 g/cm^3 at 3 and 4% dolomite content respectively.

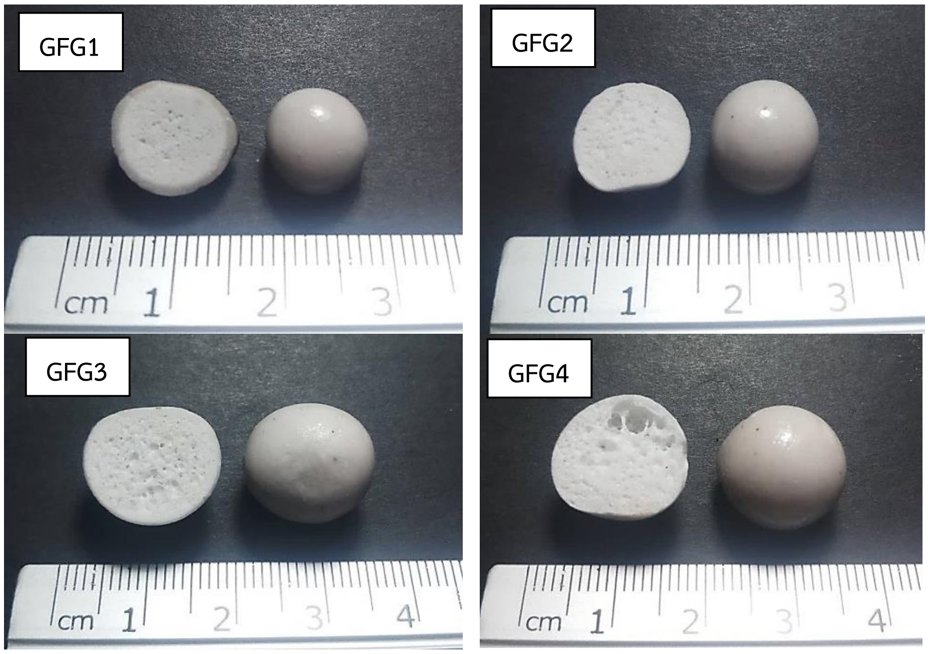


Figure 5 Effects of dolomite clay content on foaming characters of the GFGs prepared with tapioca gel binder and granulation by hand.

These GFGs were compared to those reported in the literature. The bulk density values of our GFGs produced with 3 and 4% dolomite clay conforms to the foam definition and are close to those reported previously (Konig, 2018, 11146, Arcaro, 2016, 1067). However, the use of 3% dolomite clay for the GFGs preparation gave a better uniform pore structure so it was more desirable.

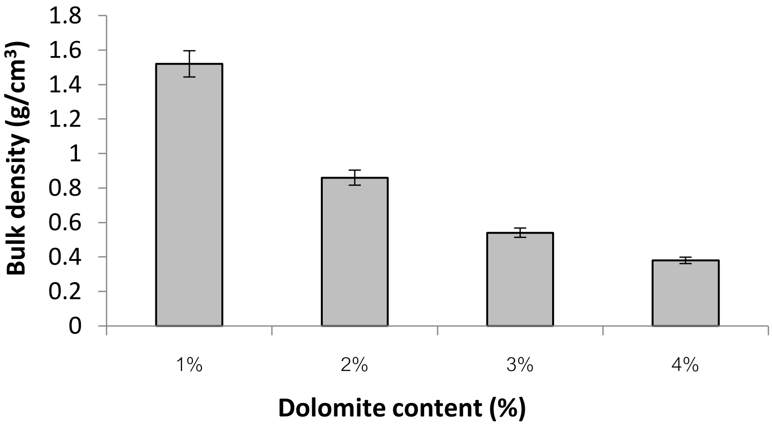


Figure 6 Effects of dolomite content on the bulk density of the GFGs prepared 35% tapioca gel and 750 °C firing.

The effect of dolomite content on true porosity, calculated according to the Eq. 3, of the GFGs prepared with tapioca gel binder is graphically displayed in Figure 7. It was shown that the only GFGs prepared with 2 - 4% dolomite clay were suitable because they exhibited porosity greater than 50%.

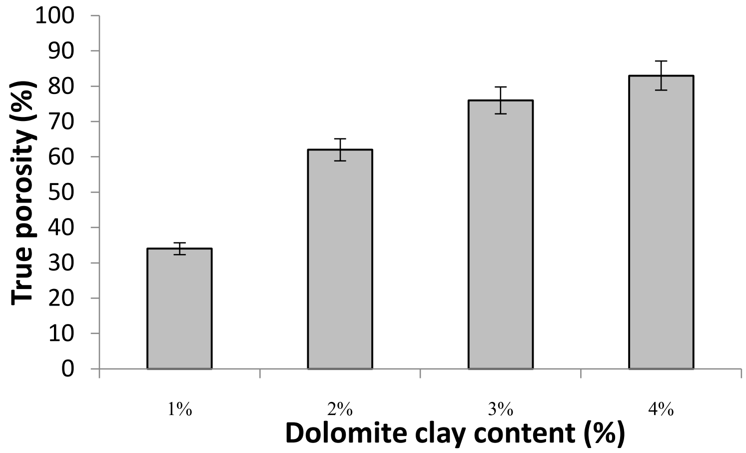


Figure 7 Effects of dolomite content on true porosity, calculated according to Eqn. 3, of the GFGs prepared 35% tapioca gel and 750 °C firing.

The pore size distribution of the GFGs prepared with 3% dolomite clay was further analyzed using Image-J software equipped to an optical microscope and its result is graphically shown in Figure 8. The main distribution of size was between 10-100 μm, with the highest number of size fell at 10 μm. This result showed that most pores were small in size and its distribution was narrow, suggesting possible use as the foamed materials. The work by Konig et al. showed that particle size of the glass cullet had a deep effect on pore size of the glass foam (Konig, 2016). The foaming was mainly caused by the reduction of manganese. When particle size of the cullet was ≤33 μm, foam glass showed a homogeneous pore distribution and a major fraction of the pores were smaller than 0.5 mm. Only when using the smallest particles (13 μm) did the pore size increase to 1 – 3 mm. due to a faster coalescence process (Konig, 2018, 11146).

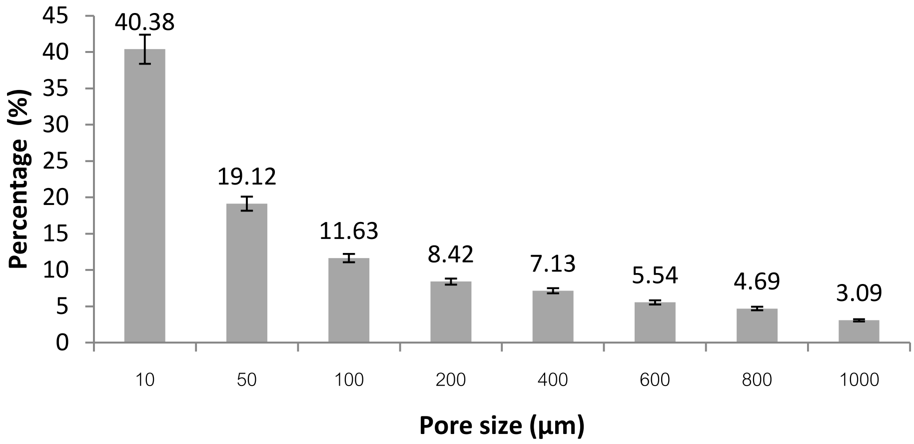


Figure 8 Pore size distribution of GFGs, as analyzed using Image J software. The GFGs were prepared with 3% dolomite clay, 35% tapioca gel and 750 °C firing.

5. Conclusions

This research showed that mixed waste glass could be used for preparation of GFGs. The proper amount of dolomite used was found to be 3% at 750 °C firing for 10 min. With this amount of the foaming agent used, GFGs had a bulk density of 0.54 g/m^3 and true porosity of 76%. Further testing of mechanical and thermal properties will be needed in order to assess quality of the prepared GFG and to incorporate it as lightweight filler in mortars. However, from literature, when the granule size was less than 10 mm, mechanical property test became difficult and was omitted (da Silva Fernandes 2019, 79). Bulk density and porosity of the GFGs reported in the literature were ranged from 0.27 g/cm^3 (Bai et al., 2014, 52), 0.46 g/cm^3 (Zhu et al., 2016, 398), 0.59-0.87 g/cm^3 (Chen et al., 2012, 3). The reported porosity values were 81.55-%(Bai et al., 2014, 52), 60-95% (Souza et al., 2017, 60), 52-62% (Chen et al., 2012, 3). The porosity and density of the GFGs prepared in this research were thus comparable to the reported values so the use as lightweight aggregates could be possible. The use of mixed waste glass to create the GFGs will not only reduce the negative environmental impact but will also create a useful material as fire-proof insulation.

6. Acknowledgements

This work was financially supported from the research funding project entitled “Utilisation of crushed glass for production of innovative eco-materials” and Mae Fah Luang University.

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