Fabrication of granular foam glass aggregates from soda lime silicate glass waste

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Abstract

Granular foam glass aggregates were fabricated from a soda lime silicate glass waste which utilized crude glycerol as a foaming agent and sodium silicate as a binder. The granulation and the foaming process were carried out by a granulator and an electric rotary furnace. The study investigated at first the foaming behavior of different foam glass formulations at various sintering temperatures. Three compositions were formulated which utilized glass powder at 85 wt%, 90 wt%, and 95 wt%, respectively. Sintering was carried out at temperatures from 800°C to 1200°C to evaluate for the foaming and melting characteristics. Foam glass with 90% glass powder showed overall uniform pore size distribution and small pores at 800°C to 900°C while heating above 1000°C resulted in sample distortion caused by glass melting and the collapse of foam glass structure. Foam glass composition with 90% glass powder was selected for the fabrication of foam glass granules at the sintering temperatures of 850°C and 900°C in rotary furnace. The best result was obtained at 850°C where the granules did not melt and stick together whereas at 900°C glass melting on the surface of the foam glass granules occurred significantly.

1. Introduction

Foam glass is a porous material in which the solid phase is mainly amorphous glass phase. The utilization of waste glass to fabricate such material can lead to various value-added products and can also help reduce waste glass disposal problems. Foam glass material consists of pore cells which can be very tiny of less than 1 mm or can be as large as few millimeters. It can be applied as an insulating material due to high porosity. In the production of foam glass, it is reasonable to use waste glasses e.g. cullet, windows and bottles [1]. The main advantages of foam glass include lightweight, low thermal conductivity, acoustic insulation, inert, non-flammable, non-toxic, good chemical and thermal stability and corrosion resistant [2-5]. Oil and gas industries utilize foam glass as insulator in pipelines. In construction industries, it has been utilized as lightweight fillers for concrete as well as exterior wall and roof [6-8].

The most promising fabrication method for foam glass is by powder processing. This method uses a fine glass powder mixed with a foaming agent or a combination of chemicals which generates gas bubbles at elevated temperature above the softening point of the parent glass. The released gas (or gases) in the softened viscous glass system leads to an increase in the bulk volume of material [9]. Foaming agents usually utilized are carbon based compounds which decompose or dissociate at high temperatures releasing CO_2/CO e.g. $CaCO_3$, Na_2CO_3, SiC, coal powder [10-12] and a combination of compounds such as MnO_2+ carbon which undergoes redox reaction [1]. Some

compounds may have not completely burnt out and lead to residue in the sample [12-14]. Factors that can affect foam glass processing by powder method include particle size, foaming agent, compaction pressure and heating conditions [13-16]. Foam glass fabricated into granular shape particles requires more processing steps than that produced into non-spherical (fractured) aggregates. It requires granulation of the powder mixture and a rotary furnace for the foaming process to prevent particle sticking due to glass melting. Granular foam glass can be advantageous when mixed with concrete mortar to produce lightweight pre-cast walls and insulating floor as it increases flowability of the concrete mix.

The objectives of this study were (1) to study the foaming behavior in soda lime silicate glass using crude glycerol as a foaming agent and sodium silicate as a fluxing agent and (2) to utilize the best foam glass composition and the most suitable foaming temperatures for the fabrication of granular foam glass aggregates.

2. Experimental

The soda-lime silicate glass waste utilized in this study was analyzed by an X-ray Fluorescence Spectrometer WDXRF ZXS Primus, Rigaku, Japan) which gave the composition in wt% as $69.2 \ SiO_2 – 12.4 Na_2O – 13.8 \ CaO – 2.4 \ MgO – 1.5 \ Al_2O_3 – 0.2 \ K_2O – 0.15 \ Fe_2O_3 – 0.02 \ P_2O_5$. The glass was ball milled in 5 kg pot mill for 6 h to obtain fine glass powder and then sieved by a Retsch AS200 sieve shaker to give < 125 µm particles. Crude glycerol (Siam Absolute Chemical Co. Ltd., Thailand)

was utilized as an effective foaming agent. Sodium silicate (Amarin Ceramics, Thailand) was used as a binder for the powder compaction and also as an effective flux in soda-lime silicate glass. The binder helps to hold the mixture composition in place during the heating process and sodium oxide goes into glass structure at above glass softening point to give non-bridging oxygens in [SiO$_4$] corner-sharing tetrahedra [16]. Solvent X was prepared in this study which consisted of 50% crude glycerol and 50% sodium silicate by volume. These two liquids were initially mixed to form homogeneous mixture and kept in an HDPE bottle for further use in foam glass preparation. The solvent X had been previously investigated by authors (unpublished data) which gave good foaming behavior at above 750°C.

The proposed foam glass compositions are shown in Table 1. Sample preparation was carried out by mixing thoroughly the glass powder and solvent X in porcelain mortar for 5 min and then pressed 10 g of the mixture into a pellet using an applied pressure of 10 MPa. The compacted green pellet had a diameter of 30 mm and ~10 mm thickness. The compacted pellet was then transferred to a chamber electric furnace for the sintering (or foaming) process. A heating rate of 5°C·min^{-1} was applied for the sintering temperatures from 800°C to 1200°C with a holding time of 10 min at maximum sintering temperature. After sintering, the sample the furnace was switched off and slow furnace cooling was allowed to avoid glass cracking.

D90 Nikon digital camera was used to capture images and to produce zoomed images for pore size analysis. The pore distribution histograms were constructed by line interception method using ImageJ® software to analyze pore size in the selected area.

The linear expansion was evaluated using simple method by measuring side view distance since for the foaming behavior the side of the sample expanded well without boundary. The cross-sectional lengths from side view also changed from top to bottom due to the curving effect in some foam glass samples. In this work, maximum side expansion length was reported by cutting the foam glass sample at half position and a vernier caliper was used to ensure precise distance.

The best composition overall in terms of small pore size, uniform pore size distribution and high sample expansion without melting was chosen for further processing into granular-shaped foam glass. The preparation method was carried out by forming compacted mixture into granular-shaped particles using a granulator. The machine had a diameter of 50 cm and the granulating process was operated at 60 rpm with an incline angle of 20º. Tiny granules of various sizes (1 mm to 3 mm) were formed and then air dried at room temperature before transferring into an electric rotary furnace for the subsequent foaming

Table 1. Foam glass composition in wt%.

Foam glass	Composition in wt%	
	Glass powder	Solvent X
FG1	95	5
FG2	90	10
FG3	85	15

process. Photographs of dynamic heating characteristics (without holding time at each measured temperature) was carried out on the compacted small pellet (2 g and 10 mm diameter) using an electric tube furnace (Carbolite, STF 16/180 UK) at a heating rate of 5°C·min^{-1}. A digital camera (Nikon D90) was used to capture the foaming behavior. This method was carried out to choose for the processing temperatures for the subsequent foaming in the rotary furnace. For the heating process in rotary furnace, a heating rate of 5°C·min^{-1} was utilized to the set maximum foaming temperature. The foaming time at maximum temperature was set to 10 min. The rotating speed for rotary heating was set at 20 rpm. When the process ended, the system was let to cool down to room temperature to obtain granular foam glass aggregates.

3. Results and discussion

In all the studied compositions, good expansion (> 50% by side expansion) occurred above 700°C. The foaming and melting characteristics of all the proposed compositions were focused on the sintering process between 800°C and 1200°C. The visual images of the sintered samples are shown in Figure 1 to Figure 3 for FG1, FG2 and FG3, respectively. Samples in FG1 to FG3 sintered at 800°C, 850°C, and 900°C resulted in excellent foaming characteristics (~50% to 60% linear expansion). The sample color became white under visual observation compared to other sintering temperatures. Cross-sectional images for FG1 and FG2 samples, sintered between 800°C and 900°C, were similar but the color became darker in FG3 due to higher amount of crude glycerol in the composition. This resulted in the incomplete combustion of carbonaceous compounds during sintering. Pore size became larger in FG3 compared to FG1 and FG2 due to the effect of high content foaming agent and flux. Foam glass samples started in shrink when sintered above 900°C and the evidence of glass melting was observed at above 1000°C from all compositions. Because of the higher the sintering temperature, the greater the melting characteristic was observed. At 1000°C, all the sintered samples in Figure 1 to Figure 3 showed non-uniform pore size as a result of large differential pressure exerted on the viscous melt during heating give pore of different sizes.
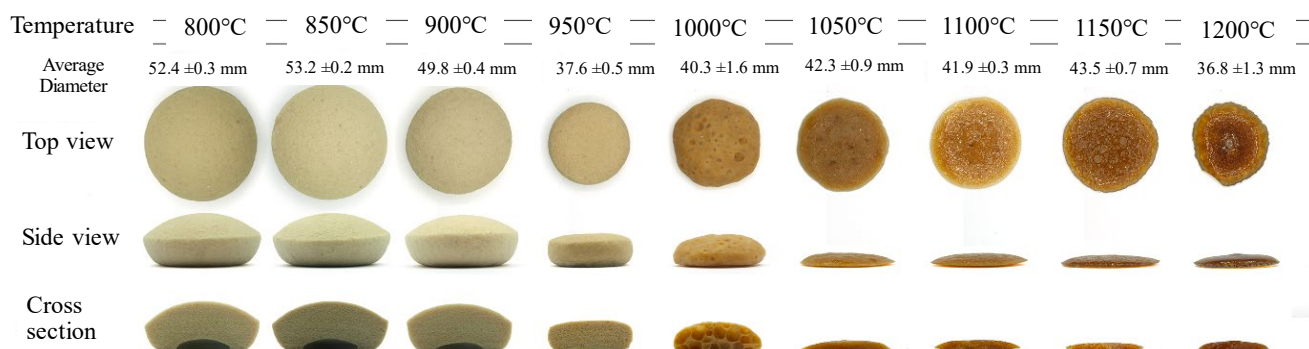

Temperature	800°C	850°C	900°C	950°C	1000°C	1050°C	1100°C	1150°C	1200°C
Average Diameter	52.4 ±0.3 mm	53.2 ±0.2 mm	49.8 ±0.4 mm	37.6 ±0.5 mm	40.3 ±1.6 mm	42.3 ±0.9 mm	41.9 ±0.3 mm	43.5 ±0.7 mm	36.8 ±1.3 mm

Figure 1. Visual observed images of FG1 composition sintered between 800°C and 1200°C.

Temperature	800°C	850°C	900°C	950°C	1000°C	1050°C	1100°C	1150°C	1200°C
Average Diameter	54.8 ±0.2 mm	55.3 ±0.4 mm	52.7 ±1.5 mm	41.5 ±0.3 mm	41.4 ±1.6 mm	39.5 ±0.8 mm	43.9 ±1.0 mm	42.0 ±1.8 mm	35.9 ±1.5 mm
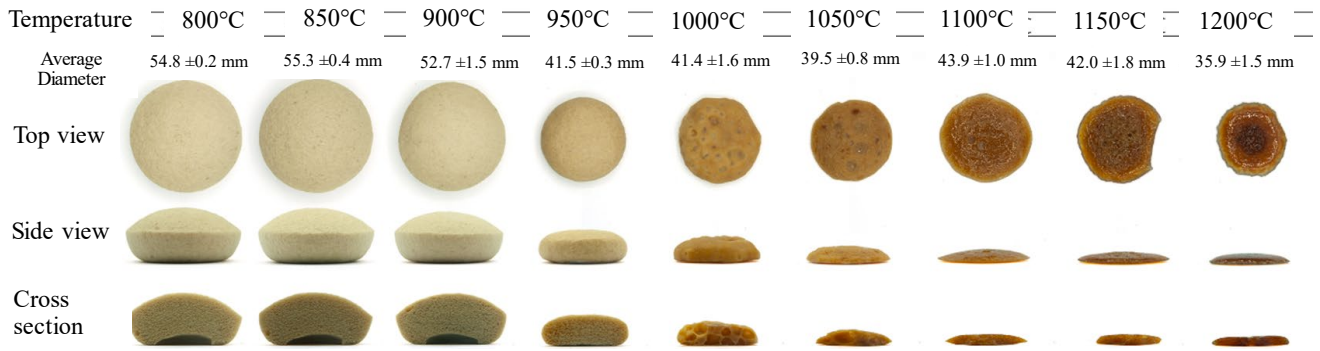
Figure 2. Visual observed images of FG2 composition sintered between 800°C and 1200°C.

Temperature	800°C	850°C	900°C	950°C	1000°C	1050°C	1100°C	1150°C	1200°C
Average Diameter	59.3 ±0.5 mm	58.3 ±0.6 mm	55.6 ±1.0 mm	44.3 ±0.8 mm	42.6 ±2.2 mm	42.3 ±1.8 mm	45.2 ±1.2 mm	43.3 ±1.5 mm	36.6 ±2.6 mm
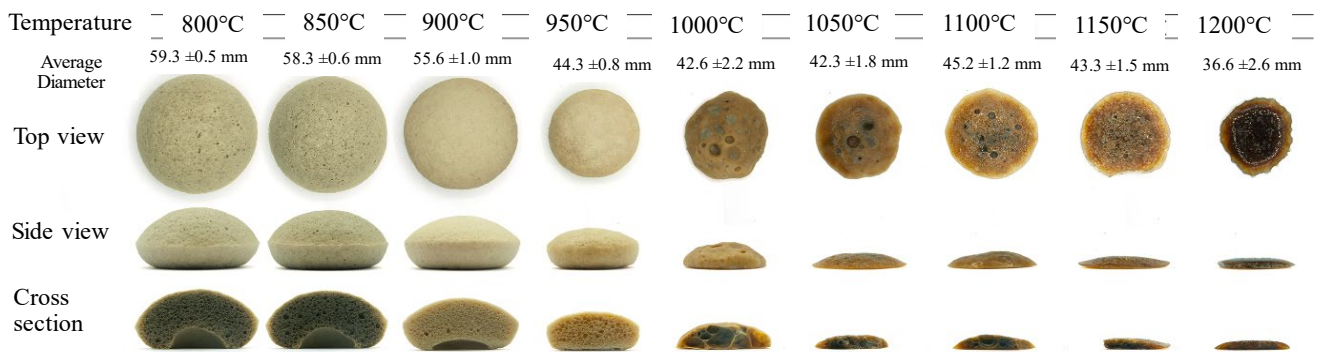
Figure 3. Visual observed images of FG3 composition sintered between 800°C and 1200°C.

Figure 4 reported average linear expansion from the maximum side view of FG1, FG2 and FG3 from the data in Figure 1 to Figure 3. For good foaming behavior by visual inspection between 800°C and 900°C it can be seen that the expansion increased with increasing crude glycerol and sodium silicate in the composition i.e. from FG1 to FG3. This occurred as a result of higher fluxing effect from sodium on the soda-lime silicate glass and the greater effect of gas release from crude glycerol decomposition. Above 900°C all the sintered samples softened and melted under their own weights.

Pore size distribution was analyzed from magnified cross-sectional images in the cropped areas. It can be seen in Figure 5 that in foam glass FG1 sintered at 800°C and 900°C, both showed a single modal distribution with similar pore size distribution. Most of the pores (>80%) are very fine and varied in the range of 200 µm to 400 µm. In FG2 samples in Figure 6, pore size distribution became broadened and more than 50% of the total pores had pore size greater than 400 µm after sintering at 800°C and 900°C. In FG3 samples (Figure 7), where the foaming agent and sodium flux were highest in all the studied compositions, it showed a significant increase in pore size at both 800°C and 900°C with more than 70% of the total pores having pore size larger than 400 µm. However, more than 15% of total pores had pore size greater than 1000 µm (or 1 mm). If this pore size range appeared in small granular samples, it would result in a non-uniform pore structure in the fabricated foam glass granules. Therefore, from linear expansion study and pore size distribution analysis, FG2 composition was chosen for further study since this composition possessed high expansion between 800°C and 900°C with small and uniform pore sizes which would be suitable for fabricating into granulated foam glass aggregates.
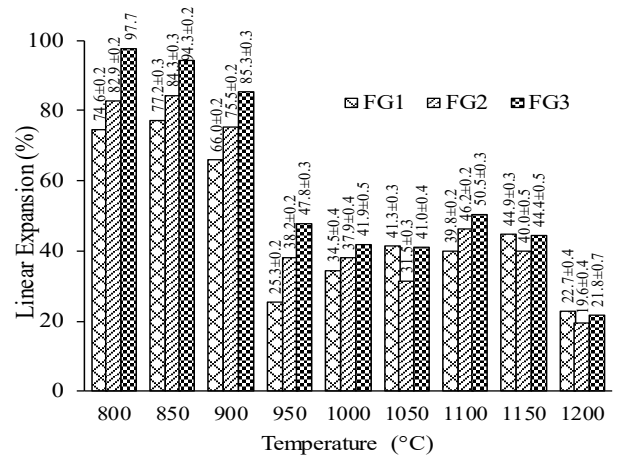
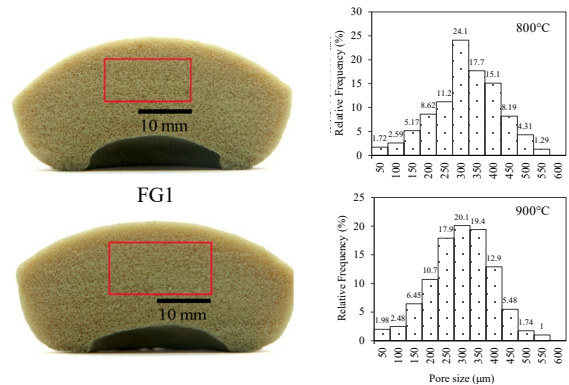**Figure 4.** Linear expansion of FG1, FG2 and FG3 sintered between 800°C and 1200°C.

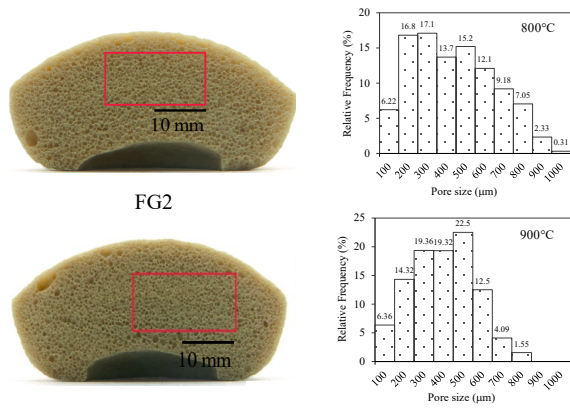**Figure 5.** Pore size distribution of foam glass FG1 sintered at 800°C and 900°C.

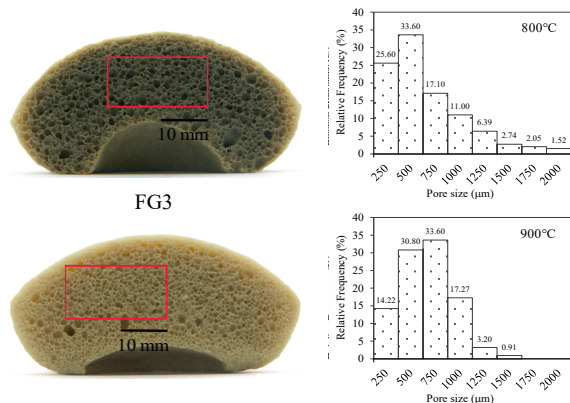**Figure 6.** Pore size distribution of foam glass FG2 sintered at 800°C and 900°C.

Figure 7. Pore size distribution of foam glass FG3 sintered at 800°C and 900°C.

Photographs of dynamic heating characteristics (without holding time at each measured temperature) on a small pellet of FG2 are shown in Figure 8. It showed similar temperature range compared with the isothermal sintering results that the highest expansion (measured by % side expansion) was obtained at 850°C and 900°C. After 900°C sample shrinkage was observed which occurred as a result of high tendency of glass melting. These two temperatures therefore were chosen for the fabrication of granular foam glass in this study.

The photographs of dynamic heating in Figure 8 when plotted for linear side expansion gave the result as shown in Figure 9. It showed a significant increase in the slope from 650°C. This temperature was related to softening point of the soda-lime silicate glass. The slope can be assumed linear with R² value of 0.98 and the straight line continued to 850°C. This equation (Y = 0.42X – 262) can be advantageous when designing the foaming process at temperatures lower than 850°C. We can then estimate from the equation for the linear expansion and possibly the expected bulk volume and density.

Figure 10 shows the overall fabrication process of granular foam glass aggregates. Granular-shaped particles (1 mm to 3 mm diameter) of FG2 composition (Figure 10(a)) were obtained after 5 min in the granulator. The rotary furnace utilized in this study is shown in Figure 10(b) in which the rotating tube was made from high alumina ceramic. The dimensions of the alumina tube of the rotary furnace were 15 mm diameter 10 mm thickness and 100 cm length (Figure 10(c)). Before the heating was conducted granular-shaped particles were filled in 1000 mL beaker and put these particles inside the rotating alumina tube then the lid was closed. The rotating speed of 20 rpm was sufficient

to fabricate into granular foam glass aggregates. Rotation was required to prevent surface sticking due to glass melting during heating. Higher rotating speed of more than 20 rpm resulted in high abrasion of compacted particles before the glass softening started to occur. This would have led to unsuccessful granular foam glass fabrication. When comparing the results from the two different heating temperatures, it can be clearly seen that heating at 900°C (Figure 10(d)) resulted in the sticking of the foam glass granules. This was related to lower glass viscosity at this temperature which melted the surface while the rotary process could not help separate the foam glass particles. Heating at 850°C, however, was successful where the granular foam glass aggregates were obtained without particle sticking. The difference of only 50°C heating temperature in the rotary furnace has led to a non-sticking effect at 850°C due to lower glass melting effect.
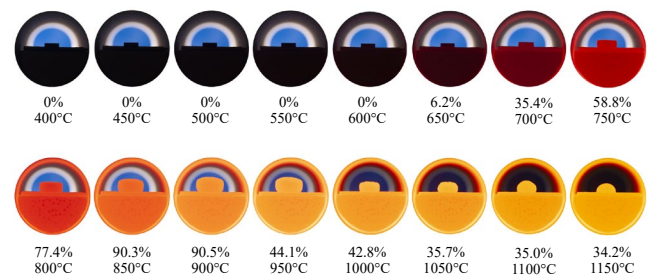

Figure 8. Expansion behavior of 10 mm diameter FG2 pellet in an electric tube furnace.
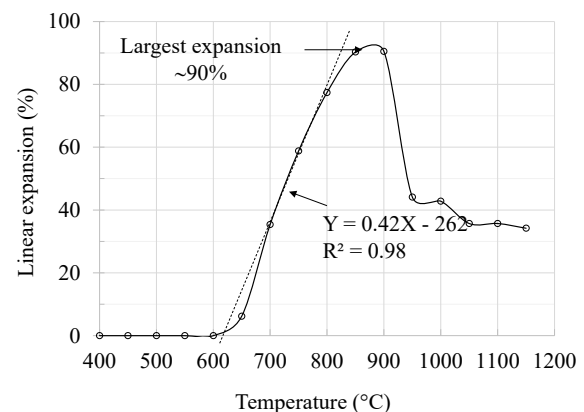

Figure 9. Linear expansion of the FG2 pellet in an electric tube furnace.
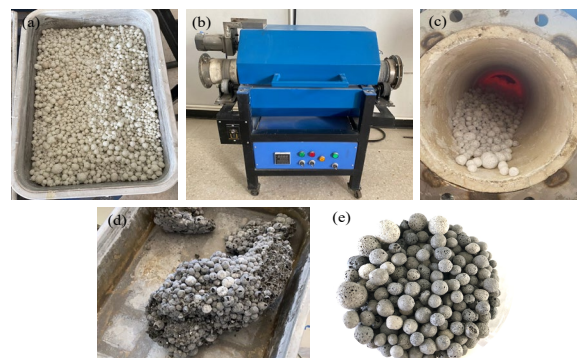

Figure 10. Fabrication process of granular foam glass aggregates (a) compacted granules of FG2 composition, (b) electric rotary furnace, (c) rotation in alumina tube, (d) processed at 900°C, and (e) processed at 850°C.

4. Conclusions

The fabrication of granular foam glass aggregates was successfully processed using an electric rotary furnace. The most suitable composition consisted of 90 wt% soda lime silicate glass and 10 wt% solvent X (50:50 by volume of crude glycerol:sodium silicate) in terms of pore structure and sample expansion. The highest bulk expansion occurred between 850°C and 900°C which was estimated from maximum side expansion (~90%). Crude glycerol gave an effective foaming characteristic by forming small closed cell pores with uniform pore size distribution. Sodium silicate was a good source of sodium flux in silicate glass and acted as a binder so that the compacted pellets and compacted granules can be easily made. Foaming in an electric rotary furnace at 850°C gave successful fabrication of granular foam glass aggregates. The foam glass particles did not stick together and were well separated. Melted foam glass particles were obtained when processed at 900°C due to high tendency of glass melting on the surface during heating even though the rotating had been continuously applied.

Acknowledgments

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