

Short communication

Glass foam from the mixture of reservoir sediment and Na_2CO_3 Yi-Chong Liao, Chi-Yen Huang^{*}*Department of Resources Engineering, National Cheng Kung University, No. 1 University Road, East District, Tainan City 701, Taiwan, ROC*

Received 15 November 2011; received in revised form 29 January 2012; accepted 30 January 2012

Available online 9 February 2012

Abstract

Glass foams were produced from recycled water reservoir sediment with 10 wt% and 15 wt% Na_2CO_3 addition, respectively, at calcining temperatures of 750–1050 °C. For the 10 wt% Na_2CO_3 samples, the pores initially formed on the surface and were sealed by the glassy phase with increasing temperature. The formation of the glassy phase was promoted by the addition of Na_2CO_3 resulting in foams being obtained at 750 °C with 15 wt% Na_2CO_3 . The increase in temperature from 750 °C to 1050 °C decreased the specific gravity (1.34 to 0.81) and the compressive strength (5.15 MPa to 1.07 MPa), which made the pores inside the foams grow and connect to each other. An excess amount of sodium ions resulted in a flowing glassy phase at high temperature, which made large pores (>1 mm) connect to each other, increasing water adsorption. The mineral phases of the foam were quartz, albite, and nepheline.

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Keywords: Sediment; Na_2CO_3 ; Foam; Bloating; Pore**1. Introduction**

Water reservoir sediment, which greatly reduces reservoir capacity, is a kind of weathering product that precipitates at the bottom of reservoirs. In order to extend the life of reservoirs, sediment is usually removed by a dredging process. Although the landfill method causes environmental problems for the downriver ecology, the swept sediment is usually buried or disposed of in situ. In Taiwan, an average amount of 14 million metric tons of sediment precipitates annually. According to a government report, the total amount of swept sediment in 2009 was 4.33 million metric tons. Recently, a method for recycling water reservoir sediment by manufacturing lightweight aggregates (LWAs), which are used to produce lightweight concrete for high-rise buildings, has been reported [1,2]. LWAs include glass foam, which has a highly porous structure that gives it the properties of acoustic and thermal insulation, and resistance to water, water vapor, and fire [3]. Many researchers have recycled wastes, such as fly ash, glass frit, and waste cathode-ray-tube glass, into glass foam [4–7]. Bernardo et al. [5] stated that the production of glass foam from wastes can

lower costs. However, few studies have discussed the production of glass foam from water reservoir sediment.

Glass foam is usually synthesized using a powder method, where glass powders are mixed with a foaming agent, such as calcium carbonate, chalk, sodium sulphate, a mixture of iron oxide with carbon, and silicon carbide [8–13]. Carbonate compounds which decompose and generate gas at lower heating temperatures are widely used as foaming agents since they are easily obtained. Foaming agents are mainly used to form a porous structure by producing gas. The formation of porous structures was discussed by Riley [14], who proposed two reasons for bloating behavior. First, the raw materials contain compounds which can form a glassy phase during heating. Second, gas is generated as the glassy phase forms, and this is then enclosed by the glassy phase, producing a porous structure.

In the traditional process for producing glass foam, fine glass powders are used as the source of the glassy phase and foaming agents are added to produce gas during the heating process. In the present study, sodium carbonate (Na_2CO_3) was mixed with reservoir sediment to produce glass foam. The sodium ions formed a glassy phase by reacting with aluminum silicates in the sediment during the heating process; the carbonate ions then decomposed to generate CO_2 gas. Therefore, glass foam can be directly produced from a mixture of sediment and sodium carbonate. The effect of the amounts of Na_2CO_3 added is discussed later in this work.

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2. Materials and methods

The reservoir sediment, with a D_{50} size of $3.2\ \mu\text{m}$, was obtained from Zengwen Reservoir, the largest in Taiwan. The sediment was first dried at $120\ ^\circ\text{C}$ for further processing. The sediment aggregate was milled into powders passed through a No. 100 mesh. The chemical composition of the sediment powders was measured using an X-ray fluorescence spectrometer (XRF, Rigaku). The Na_2CO_3 powders with a purity of 99% were passed through a No. 100 mesh. The sediment without any binders was mixed separately with 10 wt% and 15 wt% Na_2CO_3 . The weight loss was measured using a thermal analysis instrument (Netzsch) at a heating rate of $10\ ^\circ\text{C}/\text{min}$. The mixtures were pelletized into spheres (10–15 mm in diameter) by a disk pelletizer at a rotation speed of 60 rpm. In order to prevent Na_2CO_3 powders hydrating, an oleic binder was chosen to bond particles. The pelletized spheres were placed into a laboratory-scale furnace and heated for 30 min at a heating rate of $15\ ^\circ\text{C}/\text{min}$ at temperatures of 750 – $1050\ ^\circ\text{C}$. When the air inside the furnace cooled to room temperature, the foams were characterized using the procedures described below.

The bloating condition of the foam with 10 wt% Na_2CO_3 addition was observed using a heating microscope camera during the calcining process. The optical microscope was also employed to observe the macrostructures of the foams, their appearances and cross-sections. A scanning electron microscope (SEM, Hitachi TM-1000) was used to observe the microstructure of the foam with 10 wt% Na_2CO_3 addition. The Archimedes method was employed to measure the specific gravity and water adsorption after the foams had been placed in boiling water for 24 h. The specific gravity and water adsorption were calculated as follows:

$$\text{Specific gravity} = \frac{W_D}{W_S - W_I}$$

$$\text{Water adsorption (\%)} = 100 \left(\frac{W_S - W_D}{W_D} \right)$$

where W_D is the dry weight of the calcined foams, W_S is the 24-h saturated surface-dry weight, and W_I is the immersed weight in water. The compressive strength was measured using an MTS system with a crosshead speed of $0.1\ \text{mm/s}$. The compressive strength of the calcined foam was calculated using [15,16]:

$$\text{Compressive strength} = \frac{2.8P_c}{\pi X^2}$$

where P_c is the fracture load and X is the diameter of the foams. Each recorded testing value was the mean of the results from five samples. The physical properties of the foams calcined at $950\ ^\circ\text{C}$ and $1050\ ^\circ\text{C}$ were compared with those of foams produced in previous studies. Finally, powders of the sediment and of the calcined foams were prepared and passed through a No. 100 mesh. The mineral phases were measured using X-ray diffraction (XRD, Siemens D5000).

Table 1

Chemical compositions of Zengwen reservoir sediment.

Component	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O
Composition (wt%)	66.3	15.6	7.3	2.6	1.8	1.5	4.9

3. Results and discussion

Table 1 shows the chemical composition of the sediment detected by XRF. The sediment is mainly SiO_2 (66.3%), followed by Al_2O_3 (15.6%) and Fe_2O_3 (7.3%). The mineral phases of the sediment, along with their powder diffraction file (PDF) card number, are shown in Fig. 1. Mineral phases were quartz (PDF: #83-2467), dickite (PDF: #75-1484), anorthite (PDF: #89-1473), and an unknown phase. Although iron ions were not detected in the mineral phases, they may exist in the anorthite structure due to its large solid solubility. Iron oxide is also regarded as a flux for producing lightweight aggregates, since it helps the glassy phase form during the heating process. The flux oxide content was 18.1 wt% in the raw sediment, 22.6 wt% in the 10 wt% Na_2CO_3 sample, and 24.7 wt% in the 15 wt% Na_2CO_3 sample. These compositions meet the criteria for Na-glass, which means that the glassy phase can form in sediment with Na_2CO_3 added. The carbonate ions play an important role in pore formation by generating gas, leading to the porous structure.

Fig. 2 shows the thermogravimetric analysis of these samples. The weight loss of the 0 wt% Na_2CO_3 samples ranged between $450\ ^\circ\text{C}$ and $700\ ^\circ\text{C}$ due to the combustion of organic materials [17–20]. A small loss in weight at around $550\ ^\circ\text{C}$ was caused by the crystallized water from dickite, which is a clay mineral that contains crystallized water that decomposes between $500\ ^\circ\text{C}$ and $600\ ^\circ\text{C}$ [18,20,21]. The weight loss of raw sediment mainly stopped at $700\ ^\circ\text{C}$, whereas the weight loss of the samples with sodium carbonate stopped at $800\ ^\circ\text{C}$. Since sodium carbonate decomposes from $400\ ^\circ\text{C}$ to $800\ ^\circ\text{C}$, the 10 wt% Na_2CO_3 sample and the 15 wt% Na_2CO_3 sample had decomposition temperatures of $800\ ^\circ\text{C}$. The results of the thermogravimetric analysis show that weight loss increased with increasing sodium carbonate addition; the 10 wt%

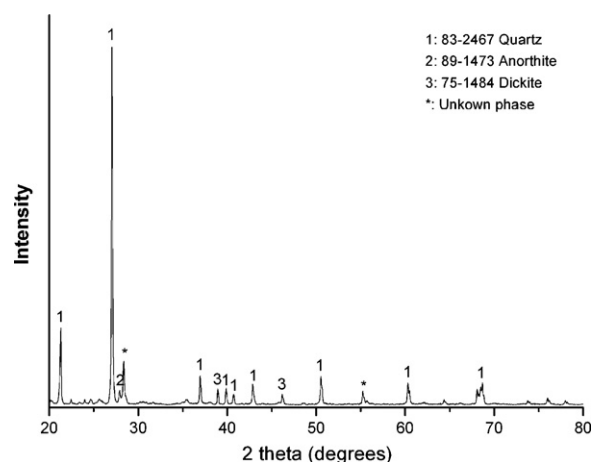


Fig. 1. Mineral phases of Zengwen reservoir sediment.

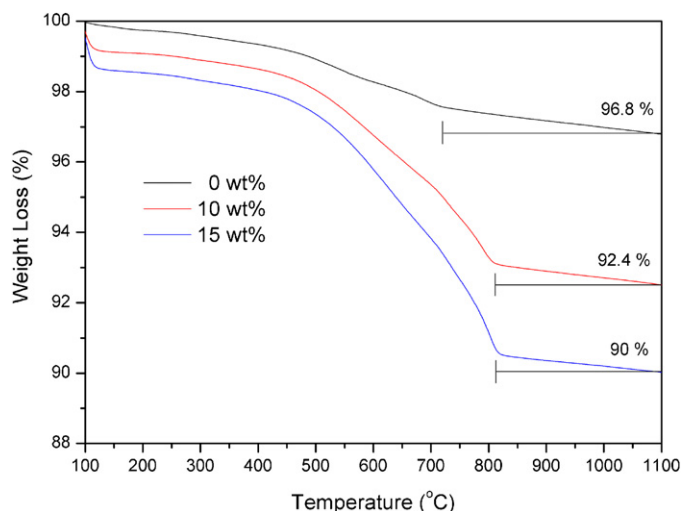


Fig. 2. Thermogravimetric analysis of samples with various amounts of Na_2CO_3 .

Na_2CO_3 sample ended around 92.4%, and the 15 wt% Na_2CO_3 sample ended around 90%. The weight difference of 2.4 wt% corresponds to the difference in the amount of sodium carbonate added to the two samples.

A heating microscope camera was employed to observe the calcining condition of the 10 wt% Na_2CO_3 sample through the window of the furnace at temperatures of 850–1050 °C. Fig. 3

shows the bloating behavior. The volume expanded and the surface became smooth with increasing temperature. At 850 °C, pores and cracks on the surface formed due to the escaping gas from the decomposition of the Na_2CO_3 and binder. At 950 °C the cracks were sealed by the glassy phase, which can promote liquid sintering between particles, although pores still existed on the surface of the foam. Some smaller pores were then sealed by the glassy phase when the temperature reached 1050 °C due to the increased viscosity. However, with increasing calcining temperature the viscosity then decreased, and larger pores started to form due to the production of gas from un-reacted Na_2CO_3 , and these were also gradually sealed by the glassy phase. This process explains why larger pores were sealed at high calcining temperature.

Fig. 4 shows the macrostructures, appearances and cross-sections of the glassy foams. It can be seen that few pores existed on the glassy surface, and that the shape of the foams calcined at high temperatures did not remain round. Pores were found around a light gray area, and their size increased with increasing temperature. The manufacturing temperature of the 15 wt% Na_2CO_3 samples was about 100 °C lower than that of the 10 wt% Na_2CO_3 samples. This indicates that sodium ions promoted the formation of the glassy phase and lowered its viscosity, explaining why pores in the 15 wt% Na_2CO_3 samples formed easily at low temperature. At low temperature, the pores exhibited good roundness. However, the pores connected to

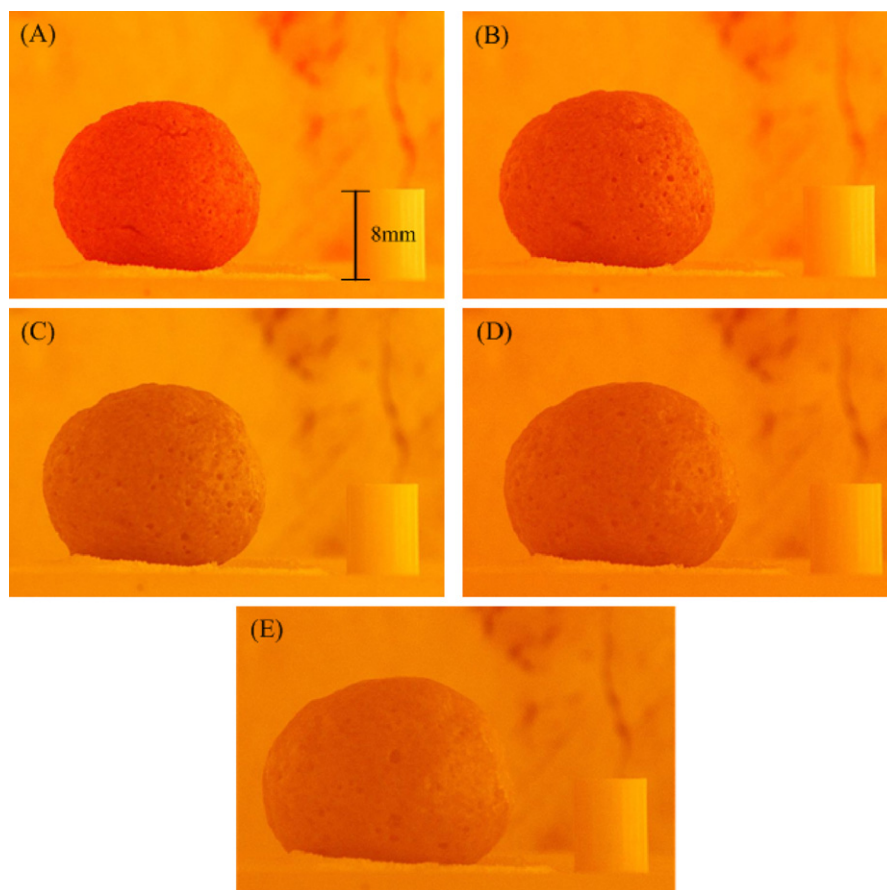


Fig. 3. Calcining conditions of the 10 wt% Na_2CO_3 sample (a) 850 °C, (b) 900 °C, (c) 950 °C, (d) 1000 °C, and (e) 1050 °C.

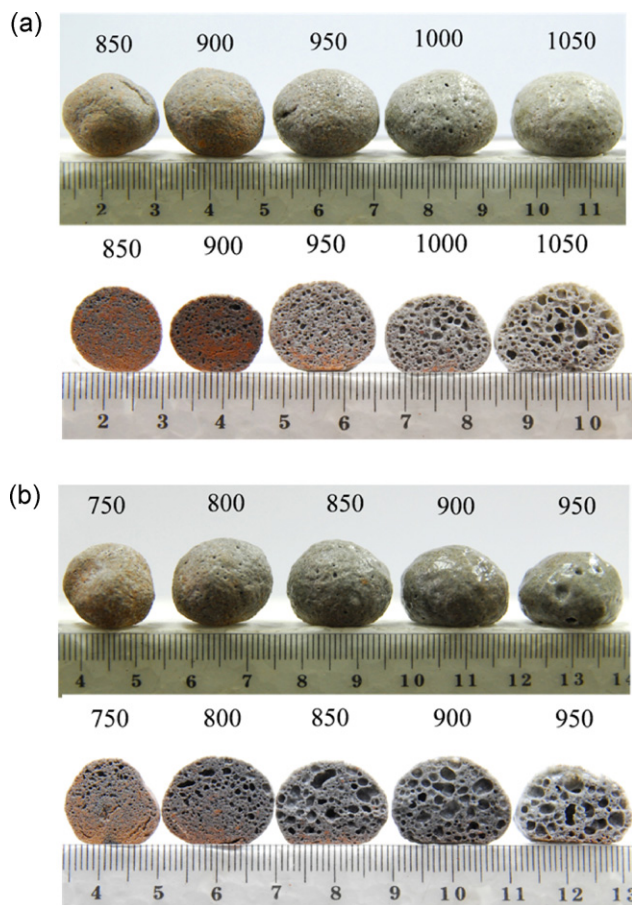


Fig. 4. Macrostructure observation of appearance and cross-section of (a) the 10wt% Na_2CO_3 samples and (b) the 15 wt% Na_2CO_3 sample.

each other and grew at higher temperature. Pores in the 10 wt% Na_2CO_3 samples were smaller than 2 mm, whereas pores in the 15 wt% Na_2CO_3 samples were larger than 3 mm. Since the addition of more Na_2CO_3 produced more gas, the pores readily became larger. Fig. 2 shows that the difference in weight loss (about 2.4%) was a result of the addition of 5 wt% Na_2CO_3 , which produced a lot of gas. Gas generation increases the internal pressure of the pores, balancing the atmosphere pressure and making the pores connect to each other in the flowing glassy phase. When the pressure is balanced between two pores, round pores start to form. This explains why pores in the 15 wt% Na_2CO_3 samples are larger than those in the 10 wt% Na_2CO_3 samples.

The physical properties of the foams are shown in Fig. 5. The compressive strengths were all higher than 1 MPa, and the specific gravities were in the range of 0.81–1.34. These results show that an increase in temperature decreased the specific gravity and the compressive strength, and the decrease of compressive strength was proportional to that of the specific gravity. The level of water adsorption, which was not positively related to the specific gravity, ranged from 14.8% to 53.7%. The water adsorption of foams with 15 wt% Na_2CO_3 addition calcined at 900 °C and 950 °C were 53.7% and 48.9%, respectively. The viscosity of the glassy phase was too low to maintain the shape of foams, and thus they started to become flat, as seen in Fig. 4(b). One reason for this is that the pores

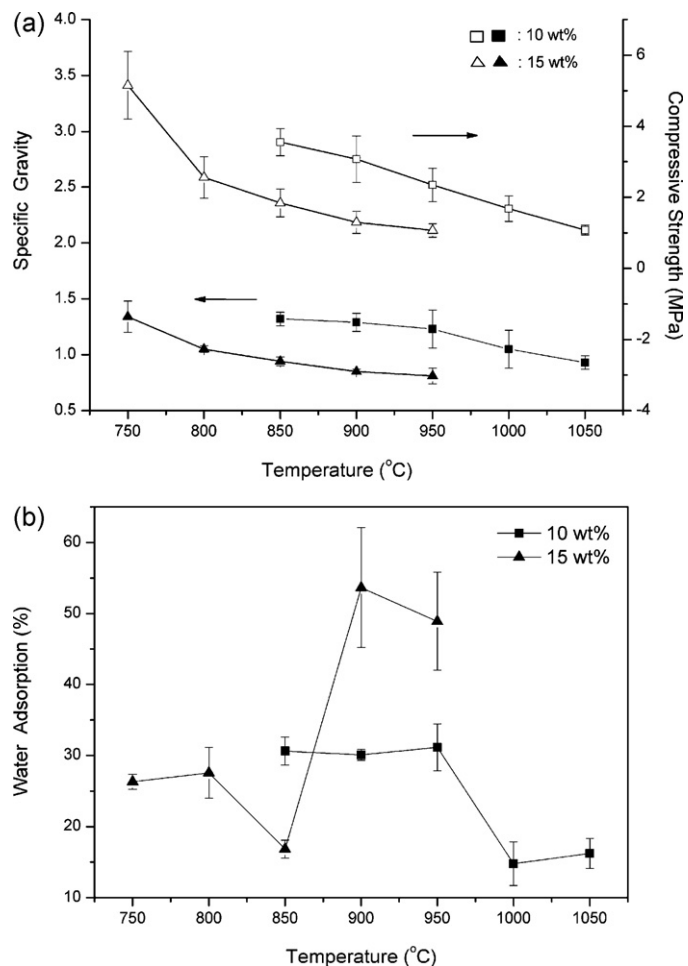


Fig. 5. Physical properties: (a) specific gravity and compressive strength and (b) water adsorption (square: 10 wt%; triangle: 15 wt%).

may readily connect to each other due to gas generation, further increasing water adsorption. Another reason may be that the samples stuck to the crucibles at high temperature, leading to a cracked surface when these samples were removed from the crucibles, and thus water penetrated them when they were tested by the Archimedes method. Table 2 lists the physical properties of the 10 wt% Na_2CO_3 sample calcined at 1050 °C and those of the 15 wt% Na_2CO_3 sample calcined at 950 °C; these samples were compared to foams produced by Bernardo and Albertini [4] and Kaz'mina et al. [22]. The results show that the foams made in this study had relatively poor physical properties and that the ratio of strength to unit weight of the particles was smaller than those found for the other foams. The process used in this study, a rolling plate granulator, was the same as that used by Kaz'mina et al. [22], while the foam produced using the pressing method (in Bernardo and Albertini [4]) exhibited the best properties.

Fig. 6 shows the SEM micrographs of the microstructure of the 10 wt% Na_2CO_3 samples. Few flat areas existed and round pores appeared at 850 °C. When the calcining temperature increased to 900 °C, smooth flat areas around the pores were observed on which there were many fine pores (under 0.1 mm). The fine pores on the flat areas were sealed by the glassy phase at 1000 °C. This explains why the water adsorption of the

Table 2

Physical properties of the 10 wt% Na_2CO_3 sample and the 15 wt% Na_2CO_3 sample compared to those of other foams.

Foam glass	10 wt%	15 wt%	Kaz'mina et al. [22]		Bernardo and Albertini [4]
	1050 °C	950 °C	Zeolite	Diabase	
Specific gravity (g/cm^3)	0.93 ± 0.06	0.81 ± 0.07	0.25	0.30	~ 0.10
Water adsorption (%)	16.23 ± 2.11	48.90 ± 6.91	<10	<10	–
Strength of particles (MPa)	1.08 ± 0.14	1.07 ± 0.19	1.80	2.00	1.00
Strength/unit weight of particles	1.16 ± 0.12	1.32 ± 0.37	7.20	6.67	10.00

10 wt% Na_2CO_3 sample decreased with increasing temperature. As for the increase in water adsorption of the 15 wt% Na_2CO_3 sample, large pores may make contact and form a new pore due to the flowing glassy phase at high temperature.

Fig. 7(a) and (b) shows the XRD patterns of the 10 wt% Na_2CO_3 sample and the 15 wt% Na_2CO_3 sample at various temperatures, respectively. The major mineral phase was quartz (PDF: #46-1045), followed by albite (PDF: #80-1094) and nepheline (PDF: #35-0424), while the anorthite phase (PDF: #89-1473) and the dickite phase (PDF: #75-1484) disappeared.

The intensity of the nepheline phase in the 10 wt% Na_2CO_3 sample weakened with increasing temperature, disappearing at 1050 °C. Albite and nepheline are Na-rich phases, and the phase transformation is caused by aluminum silicates in the sediment reacting with sodium ions from Na_2CO_3 . According to the ternary phase diagram of $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2$, the melting point locates around 750 °C when the chemical composition locates within that of these Na-rich phases. This explains why the bloating behavior occurred in the 15 wt% Na_2CO_3 sample at 750 °C.

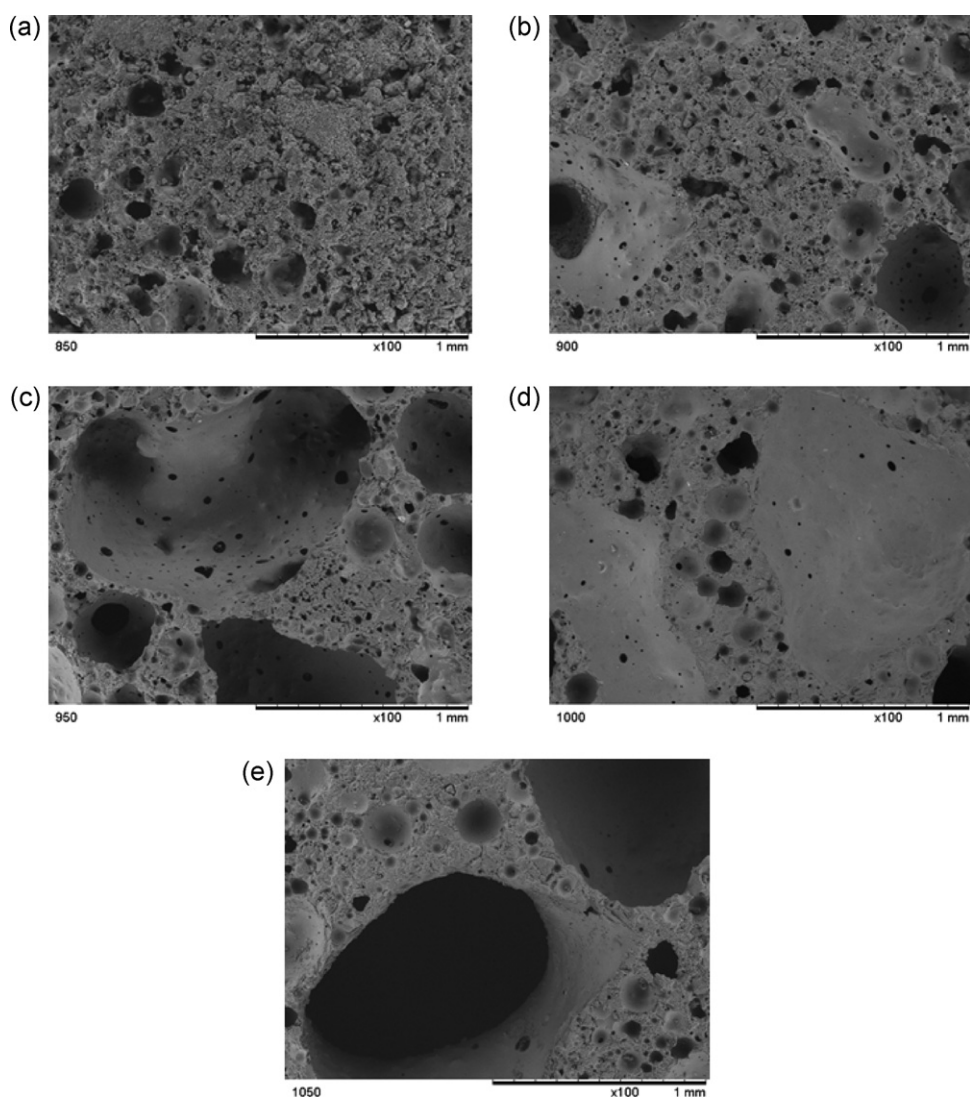


Fig. 6. Microstructure of 10 wt% Na_2CO_3 sample obtained using SEM (a) 850 °C, (b) 900 °C, (c) 950 °C, (d) 1000 °C, and (e) 1050 °C.

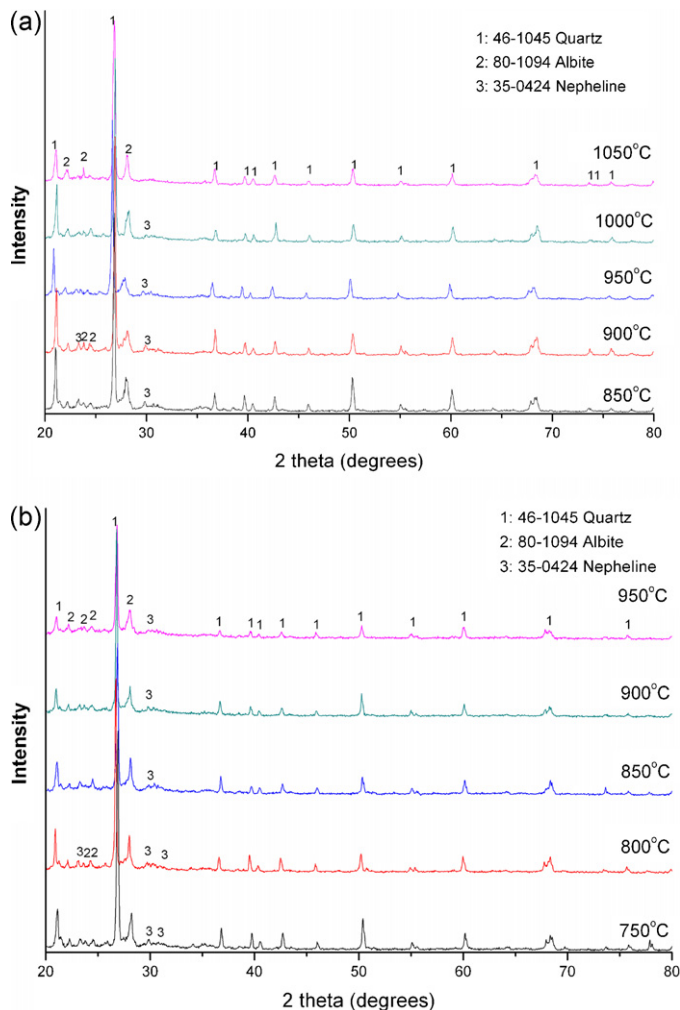


Fig. 7. Analysis of X-ray diffraction of (a) the 10 wt% Na_2CO_3 sample and (b) the 15 wt% Na_2CO_3 sample.

4. Conclusions

Water reservoir sediment can be recycled into glass foam by adding sodium carbonate. Sodium carbonate contains sodium ions which can react with SiO_2 and Al_2O_3 inside the sediment to form a glassy phase. The carbonate ions that decompose into CO_2 gas during the heating process form a porous structure. Sodium carbonate lowers the temperature for manufacturing foam, and the addition of 5 wt% Na_2CO_3 lowers the manufacturing temperature by about 100°C , with the temperature greatly affecting pore formation and pore growth. The specific gravity and the compressive strength decreased with increasing temperature, and a high temperature lowers the viscosity of the glassy phase, so the foams can not maintain good roundness. The glassy phase with a lower viscosity flows and seals the fine pores (<0.1 mm) on the internal walls of the large pores. A high temperature promotes the connection between large pores (>1 mm), which leads to an increase in the water adsorption of foams with the addition of 15 wt% Na_2CO_3 . The mineral phases of the foams were quartz, albite, and nepheline.

The physical properties of the glassy foams need to be improved if they are to be used as building materials. The

dry-pressing method presented in this work can be used to produce foams from reservoir sediment, although the pore formation and growth should be carefully controlled by adjusting the amount of Na_2CO_3 added and the temperature.

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