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Fabrication of cellular structure composite material from recycled soda-lime glass and phlogopite mica powders

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A homogeneous composite material with different physical structures has been fabricated from recycled colourless soda-lime glass powders and phlogopite-type mica powders by mixing the two powder components and sintering the mixture at a temperature above 850° C for a period of 30 min or longer. The physical structure of the composite material can be fabricated into either a cellular structure consisting of both closed and open cells or a highly densified ceramic body. The cellular structure composite material is found to have a compressive strength of about 0.877 MN m^{-2} and thermal conductivity values in the range of 0.290 to 0.306 W m^{-1} $°C^{-1}$ when measured at temperatures in the range of 25 to 100° C. The highly densified composite material, on the other hand, is found to have a compressive strength of about 53.0 MN m^{-2} and thermal conductivity values in the range of 0.198 to 0.250 W m^{-1} $°C^{-1}$. The composite material, when compared with other common building materials, is found to be potential material for construction applications because of its superior mechanical and thermal properties.

1. Introduction

Studies of thermal insulation materials in recent years have attracted a great deal of attention, because these materials play an important role in the conservation of energy. The most widely used thermal insulation materials for building insulation and other insulation applications can generally be grouped into several types, such as the rigid and semi-rigid organic plastic foams, the fibrous-type glass wool and mineral wool fibres produced from natural rocks or metallurgical slags, the loose-fill cellulose fibres produced from recycled newspapers, and the expanded mica flakes known as vermiculites. Each of these thermal insulation materials has a number of advantages as well as drawbacks depending on its application. Recent studies on solar energy collection and heat storage have found that the cellular-structure type glass or foamed glass is comparatively more suitable for solar energy work than the conventional thermal insulation materials as those mentioned above, because foamed glass is a rigid solid which has a higher structural strength and can also be used to stand for much higher temperatures.

The cellular-structure type glass or foamed glass is generally prepared by mixing glass powders of suitable composition with a gas-forming foaming agent and then reacting the mixture at elevated temperatures. Under heating, the glass powders melt, forming a viscous liquid surface envelope; and at the same time the foaming agent undergoes thermal decomposition, thereby releasing its gaseous product which causes the molten mass of the glass matrix to expand. Upon solidification, the glass mass forms a cellular structure. Although much work had been carried out by various workers in the fabrication of foamed glass, most of these studies have used one of the following three types of foaming agent, which include the carbonates of the alkali metals, such as $CaCO_3$ [1–8], carbon black [9–13] and a few metallic oxides, such as Cr_2O_3 [14], MnO_2 [15], and P_2O_5 [16]. All these foaming agents have been used effectively in producing a cellular-structure glass.

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Synthetic and natural mica powders have been used extensively in the past as electrical and thermal insulators, either alone or mixed with other materials in a composite form. Mica powders have been used as an inexpensive reinforcing agent in thermoplastics, thermoset resin, and cement boards [17, 18]. It has also been used as a filler in paints, plastics, and roofing materials [18]. Products of mica bonded with glass are considered to be an essential component in equipment for electronic, electric, and magnetic applications because of the high dielectric strength, chemical stability and mechanical properties of the glass and mica composite materials [19–21].

The present paper describes a fabrication process which could produce a homogeneous composite material with different physical structures from recycled colourless soda-lime glass powders and a locally produced phlogopite-type mica powder. A cellular structure of the glass-mica system has been fabricated without the use of a gas-forming foaming agent, as the mica powders themselves act as a foaming agent. The fabricated products are found to have good thermal insulation values and also superior mechanical properties, as compared with other foamed glass products prepared from gas-forming foaming agents, and with other common building materials.

2. Experimental procedure

The glass powders used for the present investigation were prepared from recycled colourless soda-lime glass cullets.[*] The glass cullets were first pulverized into fine grains (in the range of 0.2 to 0.5 mm diameter) by a mechanical pulverizer. The pulverized glass grains were then ground into fine powders by a dry ball-milling grinding process for a period of about 5 h. The corundum grinding media were used. The glass powders used for sample preparation were mostly in the 150 to 300 μm range. Chemical analysis showed that the recycled glass powders consist of the following compositions

Components	*Wt %*
SiO_2	71.30
Na_2O	11.30
CaO	8.39
Al_2O_3	2.24
MgO	0.09
Fe_2O_3	0.43
Other oxides	6.25

The mica powders used for the investigation were of the phlogopite type and were obtained from a local producer.[†] The mica powders used for sample preparation were finer than 300 μm, and their chemical composition, as reported by the producer, is known to be as follows [22]

Components	*Wt %*
SiO_2	40.7
Al_2O_3	15.8
MgO	20.6
K_2O	10.0
FeO	7.8
F	2.2
Fe_2O_3	1.2
BaO	0.5
H_2O	1.0

For most experimental evaluation purposes, a 50 g specimen was prepared from appropriate portions of the recycled glass powders and mica powders. The weighed amount of each powder was thoroughly mixed in an agate mortar and pestle. In order to improve the particle adhesion of the powder mixture, 3 ml tap water was added to the powders during the mixing operation. The mixed powders were then compressed into a circular disc measuring about 50 mm diameter and 20 mm thick, employing a compaction pressure of about 1 MN m^{-2}. The compressed powder compact was then placed on a refractory brick and sintered in a Glo-Bar electric heating furnace at atmospheric conditions. A wide temperature range varying from 780 to 900° C and a soaking period varying from 15 to 120 min have been used, but a combination of 850° C and 30 min soaking were found to produce the most satisfactory results. The temperature of the Glo-Bar furnace was raised to the selected level steadily with a heating rate of approximately 300° C h^{-1}. After soaking at the selected temperature level for the desired period, the furnace was rapidly cooled down to about 600° C and equilibrated at that temperature for

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[*] Supplied by a local glass manufacturer, Consumer Glass Ltd. Montreal, Canada.

[†] The phlogopite-type mica is processed in Boucherville, Quebec (near Montreal) from a high purity ore, by the Marietta Resources International Ltd, in joint venture with Societe Mineralurique Laviolette, Inc, Montreal, under a trade name known as "Suzorite" mica.

about 30 min before further cooling down to about 300° C at a slow cooling rate. The total time required to complete the cooling cycle before the removal of the sintered specimen from the furnace was about 4 h.

The physical properties of the sintered specimen were characterized by measuring the bulk density, the apparent porosity, the water absorption, and the volumetric change according to the procedure described in the ASTM-C20 methods [23]. The compressive strength of the sintered composite compacts were measured with an Instron Universal Testing Instrument, Model 1125; and the thermal conductivity of the prepared samples was determined with a Dynatech Guarded Hot-Plate according to the testing procedure described in the ASTM-C177 method [24], and based on a larger specimen measuring 20 cm diameter.

3. Results and discussion

In the present study, compositions ranging from 95 wt % glass + 5 wt % mica to 25 wt % glass + 75 wt % mica have been examined. The sintered glass–mica mixtures were found to be a homogeneous composite material with the mica particles uniformly distributed in the solidified glass. Most of the sintered samples containing a large proportion of glass are rigid and hard ceramic bodies of amber-brown colour. Bulk density measurements showed that the bulk density of the sintered composite compacts depends strongly on the composition of the mixture. Fig. 1 shows the variation of the bulk density with mica content. It is evident that a complex relationship exists. When 5 wt % mica was present in the mixture, the sintered composite compact exhibited a relatively high bulk density value, about $1.72 \times 10^3 \mathrm{kg\,m^{-3}}$, which is lower than the density value of the recycled soda-lime glass powders, about 2.46×10^3 $\mathrm{kg\,m^{-3}}$. As the mica content in the mixture increased, the bulk density of the sintered composite compacts decreased gradually and reached a minimum value of about $0.75 \times 10^3 \mathrm{kg}$ $\mathrm{m^{-3}}$ when the mixture contained about 10 wt % mica. As the mica content in the mixture further

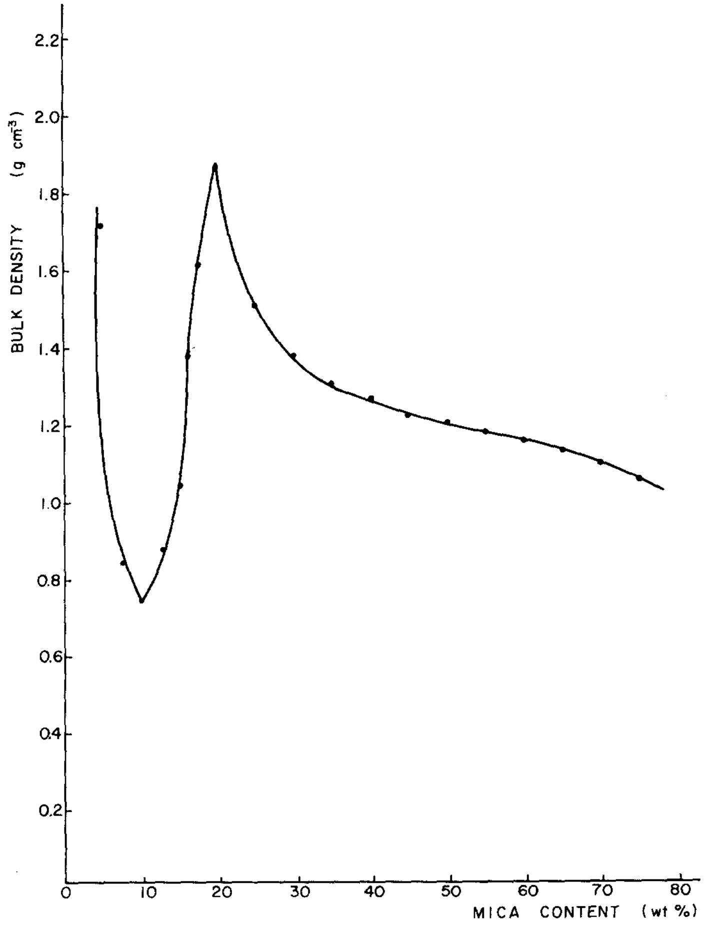

Figure 1 Variation of bulk density with mica content in the sintered composite compacts of recycled colourless soda-lime glass and phlogopite mica powders.

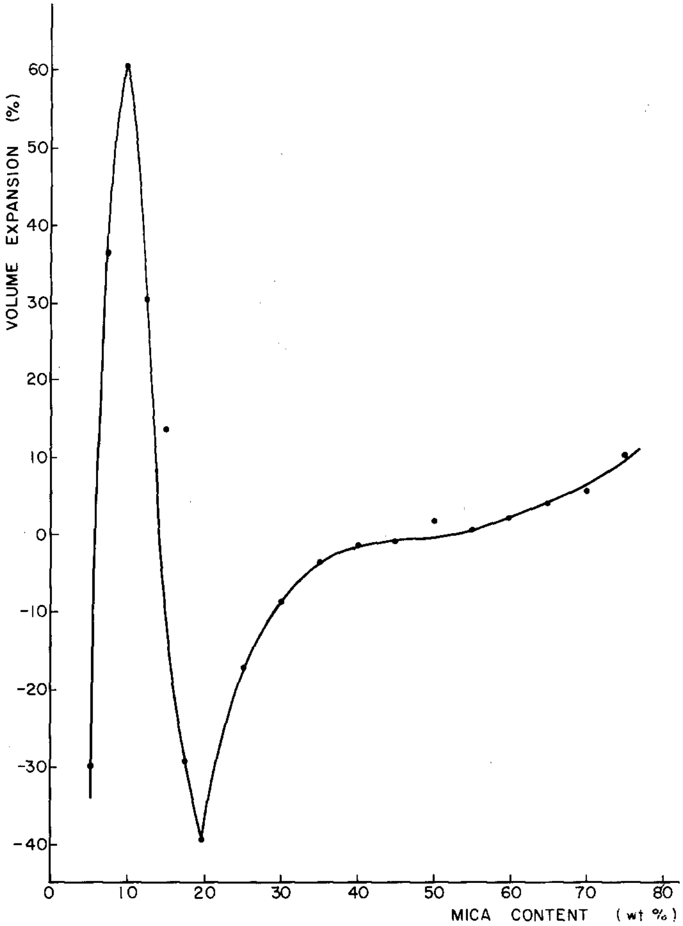

Figure 2 Variation of volume expansion with mica content in the sintered composite compacts of recycled colourless soda-lime glass and phlogopite mica powders.

increased, the bulk density of the sintered composite compacts began to rise and a maximum value of $1.875 \times 10^3 \mathrm{kg\,m^{-3}}$ was attained when 20 wt% mica was present in the mixture. Further increase of mica content in the mixture beyond this maximum limit, the bulk density of the sintered composite compacts gradually decreased up to 75 wt% mica content.

Measurements of the associated volume changes of the sintered composite compacts before and after sintering treatment also showed a similar unusual behaviour, but in a reverse direction. Fig. 2 shows a plot of the volume expansion (taken as the ratio of the difference between the final volume and the initial volume to the initial volume of the powder compact) with the mica content. It is evident that a volume expansion of about 60% was attained when 10 wt% mica was present in the composite mixture and a maximum shrinkage of about 40% was observed when 20 wt% mica was present in the mixture. Results of both bulk density and volume expansion measurements have definitely indicated that a structural modification has taken place in the glass–mica matrix.

The existence of a structural modification in the glass–mica matrix was substantiated by examining the micro-structure of the sintered composite compacts. When more than 7.5 wt% and up to 15.0 wt% mica powders was present in the mixture, the sintered compact showed a cellular structure with both open and closed cells, measuring 0.2 mm or smaller in size. However, when about 10 wt% mica powders were present in the mixture, the cellular structure developed in the sintered compact consists of most closed cells with sizes ranging from 0.4 mm to as large as 7 mm. When the mica content in the mixture was more than 15.0 wt% and up to 20 wt%, the cellular structure no longer existed in the sintered compact. Fig. 3 shows a photomicrograph of a glass–mica composite compact composed of 10 wt% mica and 90 wt% glass, and sintered at 850° C for 30 min. The cellular structure is obvious, and its appearance was found to be very similar to the cellular struc-

Figure 3 Photomicrograph of a typical sintered composite compact composed of 10 wt% mica and 90 wt% colourless glass and sintered at 850° C for 30 min.

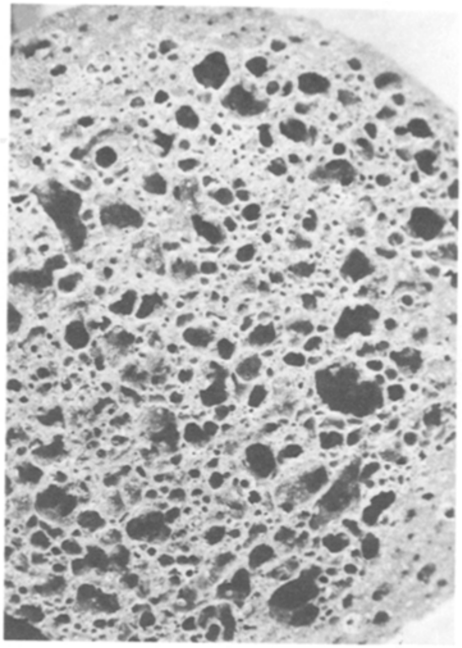

Figure 4 Photomicrograph of the cellular structure of a typical recycled colourless soda-lime glass sample formed with sodium carbonate (Na_2CO_3) as gas-forming foaming agent.

ture developed in the same recycled soda-lime glass molten matrix foamed with the use of sodium carbonate (Na_2CO_3) as the gas-forming foaming agent, as shown in Fig. 4.

The development of a cellular structure in the sintered glass–mica mixture within the narrow range of mica content of 7.5 to 15.0 wt% can probably be explained by the following reasoning: as the sintering temperature was raised to the 850° C level, most of the glass grains near the surface of the powder compact would be melted, and the melting process gradually propagated to the interior of the compact. At this elevated temperature, the mica particles are known to undergo no thermal decomposition or to cause any physical changes; however, the mica particles do undergo a dehydration process, thereby releasing both its free water and combined water [22]. Since the glass melting process is believed to progress from the surface to the interior, the viscous liquid glass on the surface region will, therefore, form a closed envelope. Because of the presence of this liquid envelope, the released water vapour cannot escape to the outer region of the molten matrix, and as a result, the entrapped water vapour developed a strong vapour pressure which consequently pushed the molten glass mass apart, or caused the glass mass to expand. As the liquid glass–mica is cooled suddenly, solidification of the composite matrix takes place with the formation of a cellular structure.

The absence of a cellular structure in the sintered composite compact when mixtures contained more than 15.0 wt% mica particles, and the attainment of a maximum densification in the sintered compact when 20 wt% mica was present in the composition, can probably be assessed using the following arguments: as the mica content in the mixture is increased, or the glass content is decreased, the molten mass of the glass at elevated temperature became less available to wet all the mica particles, as well as to form a tight viscous liquid envelope for entrapping the water vapour which is released from the dehydration process of the mica particles. Because of the lack of development of a strong vapour pressure to expand the molten glass, the volume expansion is, therefore, weakened. When the mica content in the mixture approaches 20 wt%, it is quite possible that the large number of mica particles which are thoroughly wetted and submerged in the molten glass mass are balanced out with the viscous glass mass and particle bridging, or particle interlocking is absent. As the molten composite matrix solidifies in the

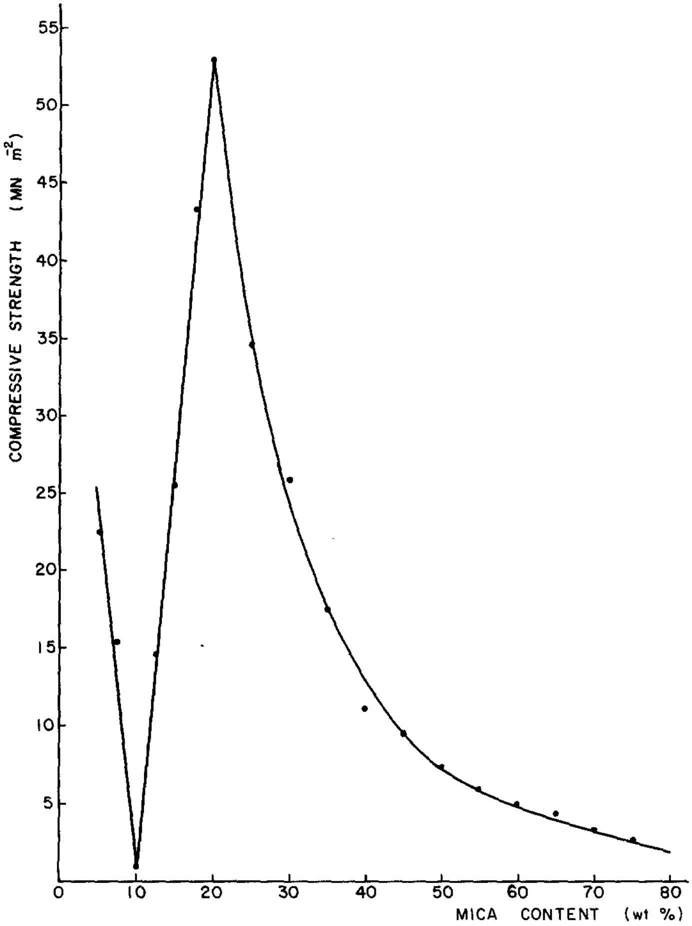

Figure 5 Variation of compressive strength with mica content in the sintered composite compacts of recycled colourless soda-lime glass and phlogopite mica powders.

cooling process, it contracts to the extent of the glass component alone, as if the mica particles were absent. It is this extensive solidification of the glass that contributes to the maximum densification.

The gradual decrease in bulk density of the sintered composite compacts beyond the maximum densification at 20 wt % mica content as that shown in Fig. 1, can probably be attributed to the significant difference of the density of the glass powders ($2.46 \times 10^3 \mathrm{kg\,m^{-3}}$) and the bulk density of the mica particles ($0.28 \times 10^3 \mathrm{kg\,m^{-3}}$) [22]. As the mica content is increased beyond the 20 wt % range, the total number of mica particles also increases in the mixture thereby contributing to a lower bulk density. It is also due to the presence of this large number of mica particles in the mixture, that the particle bridging or interlocking effect also increased, thus causing a small degree of volume expansion upon solidification of the mixture, as that shown in Fig. 2.

The compressive strength of the sintered glass–mica composite compacts was found to depend strongly on the amount of the glass powders which were present in the mixtures. When the compressive strength of the sintered composite compact was plotted as a function of the mica content, a non-symmetrical variation was again obtained, as shown in Fig. 5. It is evident that the compressive strength of the sintered compact has the lowest value of about 0.877 MN m^{-2} when 10 wt % mica was present in the composite mixture and the highest value of about 53.0 MN m^{-2} when 20 wt % mica was present in the mixture. A striking resemblance exists between Fig. 1 and Fig. 5 for its dependence on the mica content. Fig. 6 shows the dependence of the compressive strength on the bulk density of the sintered glass–mica composite mixtures. Data plotted as open circles were obtained from samples with less than 20 wt % mica in the mixture and data plotted as solid dots were obtained from samples with more than 20 wt % mica in the mixture. It is evident that a near linear relationship exists between bulk density and compressive strength, particularly for samples with high bulk density values.

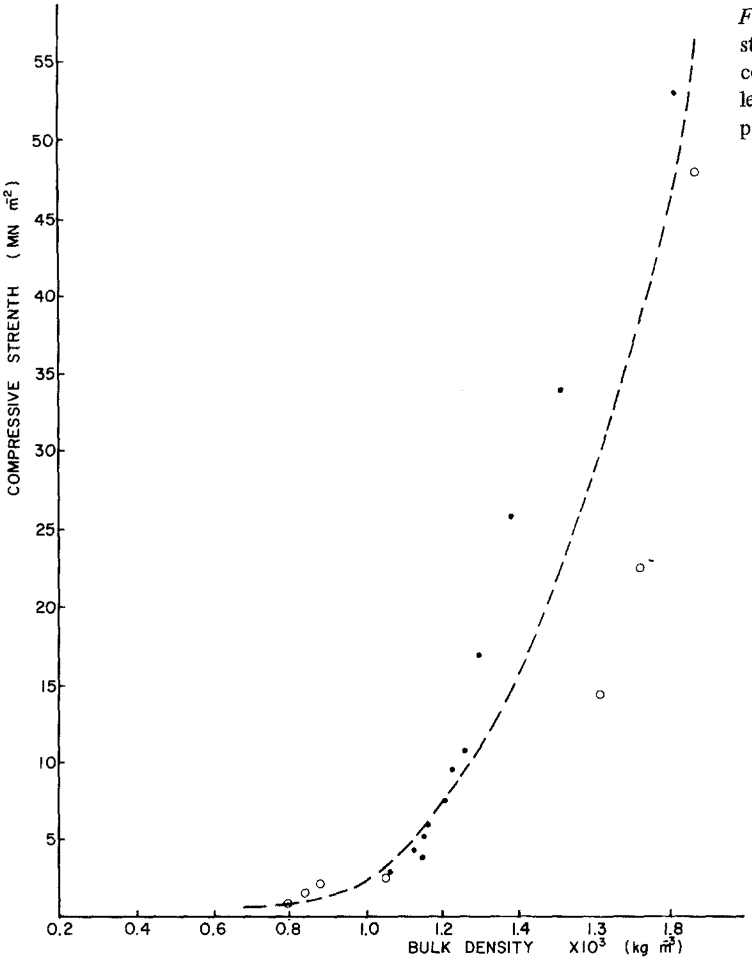

Figure 6 Relationship of compressive strength and bulk density of sintered composite compacts of recycled colourless soda-lime glass and phlogopite mica powders.

The average compressive strength of the cellular structures composite compacts of glass and mica mixtures determined from a number of similar test samples was found to be about 0.874 MN m^{-2}, which is slightly higher than similar cellular structure glass which is prepared from gas-forming foaming agent. For example, when the present recycled colourless soda-lime glass powders were foamed with calcium carbonate ($CaCO_3$) as foaming agent, a bulk density of about 0.25 x 10^3 kg m^{-3} and a compressive strength of about 0.462 MN m^{-2} were obtained. Similarly, when these glass powders were foamed with sodium carbonate (Na_2CO_3) as foaming agent, a bulk density of about 0.62 x 10^3 kg m^{-3} and a compressive strength of about 0.744 MN m^{-2} were obtained. The compressive strength (53.0 MN m^{-2}) of the highly densified glass–mica composite mixtures was found comparable to some of the common building materials, such as common building bricks, concrete, and cement mortar.

The thermal conductivity of the sintered glass–mica composite mixtures was found to vary slightly with both the composition of the mixture and the temperature at which the measurements were made. The thermal conductivity of the glass–mica composite material having the cellular structure was found to be in the range 0.290 to 0.306 W m^{-1} °C^{-1} when measured at the temperature range of 25 to 110° C. These thermal conductivity values are very similar to those observed in the cellular glasses prepared from carbonate foaming agents. On the other hand, sintered glass–mica composite mixtures having a highly densified ceramic body have a thermal conductivity value in the range of 0.198 to 0.250 W m^{-1} °C^{-1} when measured in the same temperature range. The thermal resistance of the sintered glass–mica composite material of the latter type is, therefore, much higher when compared with that of some of the common building materials, such as common bricks, concrete, cement mortar, and solid soda-lime glass sheets. The experimental results of the thermal conductivity values of the glass–mica composite materials,

TABLE I Thermal conductivity values of sintered glass–mica composite materials and other related building materials at various temperatures

Description of materials	Temperature (°C)	Thermal conductivity ($\mathrm{W\,m^{-1}\,°C^{-1}}$)
90 wt% glass + 10 wt% mica cellular structure composite	25–30	0.290
	50–60	0.299
	80–90	0.306
80 wt% glass + 20 wt% mica highly densified composite	25–30	0.194
	50–60	0.204
	100–110	0.250
50 wt% glass + 50 wt% mica moderately bonded composite	25–30	0.193
	50–60	0.197
	100–110	0.209
Cellular structure soda-lime glass foamed with Na_2CO_3 foaming agent	30–40	0.245
	60–75	0.272
	110–120	0.282
Common building brick*		0.719
Fireclay brick*		1.004
Soda-lime glass sheet*		1.021
Cement mortar*		0.719

*These data are quoted from the ASHRAE Handbook of Fundamentals, Chapter 22, 1977.

and a few other related building materials can be summarized, as shown in Table I.

4. Conclusions

The present experimental study has demonstrated that recycled soda-lime glass powders and phlogopite-type mica powders can be mixed together and fabricated into a homogeneous composite material by sintering at elevated temperatures and that the physical structure of the composite material can be altered by controlling the composition of the two starting components. When mica powders in the range of 7.5 to 15.0 wt% are mixed and reacted with the glass powders, a cellular structure with a maximum volume expansion is attained when 10 wt% mica is present in the powder mixture. This cellular structure composite material has an average compressive strength of about $0.877\,\mathrm{MN\,m^{-2}}$ and a thermal conductivity value in the range 0.290 to $0.306\,\mathrm{W\,m^{-1}\,°C^{-1}}$ when measured in the temperature range 25 to 100° C. Both mechanical and thermal properties of the cellular structure glass–mica composite material are found to be superior when compared with other cellular structure glasses which are prepared from the use of carbonate-type foaming agents. On the other hand, when a mica powder content greater than 15 wt% is mixed and reacted with the glass powders, a highly densified ceramic body is obtained and a maximum densification of the glass–mica system is attained when 20 wt% mica is present in the mixture. The densified glass–mica composite material has a compressive strength greater than $53.0\,\mathrm{MN\,m^{-2}}$, which is comparable to some of the common building materials, such as common building bricks, concrete and cement mortar. The thermal conductivity of the densified glass–mica composite material is in the range of 0.198 to $0.250\,\mathrm{W\,m^{-1}\,°C^{-1}}$ when measured at the temperature range of 25 to 100° C. The thermal resistance values of the glass–mica composite material is, therefore, considerably higher than that of some of the building materials mentioned above. The possession of superior mechanical strength and high thermal resistance suggests that the glass–mica composite material can be employed for construction applications because of its energy conservation potentials.

The formation of the cellular structure in the narrow range of compositional variations is probably attributed to the dehydration of the mica particles at elevated temperatures and the entrapment of the released water vapour in the viscous liquid glass. Because of the viscous nature of the molten glass, the entrapped water vapour developed a strong vapour pressure which causes the molten glass to expand in the continuous heating process. Upon sudden cooling, the liquid glass–mica matrix becomes solidified, thus forming the cellular structure. On the other hand, the formation of a highly densified ceramic body of the composite material at the specific composition of the mixture, namely 80 wt% glass and 20 wt% mica, is probably attributed to the absence of an expanding force in the viscous liquid glass–mica matrix and also to the equilibrium balance of the liqud glass phase and the solid mica phase. It is conceived that when more mica particles are present in the mixture, the liquid glass phase becomes insufficient to form a closed envelope to prevent the escape of the water vapour which is released from the mica particles during the dehydration process. Instead, the released water vapour from the mica particles gradually escapes from the viscous liquid glass-mica matrix, thereby developing no strong vapour pressure to expand the liquid phase. It is also postulated that perhaps there exists no particle interlocking or particle bridging among the solid mica particles during the solidification process, thus allowing a maximum densification to occur.

It is believed that this publication is the first piece of evidence demonstrating the formation of a cellular structure in the glass–mica system without the use of a gas-forming foaming agent. Further experimental work is in progress to explore the full potential of the composite material, particularly for construction applications. Results of these studies will be published in future papers.

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