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The effects of temperature, pressure and water on the preparation of glass-mica composite material

NORMAN M. P. LOW
Centre for Building Studies, Concordia University, Montreal, Canada

A systematic investigation on the fabrication of glass–mica composite materials from recycled colourless soda–lime glass powders and phlogopite-type mica powders has been conducted. Mixtures of two specific compositions of the glass–mica system were used and the investigation was based on several chosen processing parameters. When compacted powder samples were sintered at temperatures in the range 780 to 900° C, samples of one composition formed a composite material having a cellular structure; whereas samples of the other composition formed a composite material having a highly-dense ceramic structure. Sample evaluations showed that both the sintering temperature and the quantity of water which is added to the glass–mica mixtures as wetting agent in the powder compaction process are sensitive processing parameters. They can control the physical, mechanical and thermal properties of the glass–mica composite material. It was found that when glass–mica dry mixtures were prepared with the addition of a quantity of water equivalent to about 10% of the sample weight and sintered at the temperature of 850° C, the resultant composite material exhibited optimum physical, mechanical and thermal properties. The compressive strength and thermal insulating value of the glass–mica composite material with the densified structure are found to be superior to those of several conventional building materials, such as masonary products, lightweight concrete and soda–lime glass components. The experimental findings suggest that the glass–mica composite material is a potential structural element for building construction applications as it may contribute to energy conservation.

1. Introduction

In a previous paper [1], it was shown that when a mixture of recycled soda–lime glass powder and locally produced phlogopite-type mica powders were compressed into powder compacts and sintered at elevated temperatures in the range of 780 to 900° C in air, composite materials of the mica–glass system with different physical structures were produced. By close control of the composition of the two starting components, it was found to be possible to change the physical structure of the mica–glass composite material from a cellular structure consisting of both closed cells and open cells to a highly densified ceramic body with very little porosity. Experimental results exhibited a complex relationship between the bulk density and volume expansion of the sintered samples with mica content. Evaluation showed that the mica–glass composite material having the cellular structure has a relatively low compressive strength but a high thermal resistance which is compatible with other thermal insulation materials consisting of similar cellular structure. On the other hand, mica–glass composite material having the densified ceramic body was found to have a comparatively high compressive strength and superior thermal resistance compared with other conventional building materials, such as building clay bricks, lightweight aggregate concrete and cement mortar. The mica–glass composite materials appear to have significant potentials as building material.

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Although glass-bonded mica has been fabricated and used in the past as a good dielectric material for industrial applications [2–4], very little information concerning the material preparation conditions and their effects on the properties of the composite material has been reported in the literature. As a general rule, in the glass-bonded mica fabrication processing [2], it has been reported that a higher percentage of mica is used in the compressive moulding fabrication process and a lower percentage is used in the transfer or injection moulding fabrication process. Typically, the formulation and preparation used for compression moulding is a composition consisting of 60 wt% mica and 40 wt% glass and for the transfer moulding is the use of a composition consisting of 40 wt% mica and 60 wt% glass. As shown in the previous study [1], glass–mica structures consisting of 40–60 wt% mica exhibited very little variation in physical properties with compositional change within this narrow range. This can be illustrated with Fig. 1, which shows the variation of specimen volume of the glass–mica mixtures after sintering treatment at 850° C for 30 min as a function of mica content in the mixture. It is evident that the dimensional change of the sintered specimens which contained 40–60 wt% mica is virtually negligible.

Densification of ceramic materials by a solid-state sintering process is often influenced by several processing variables, such as particle packing, sintering temperature and particle bonding agent. The glass–mica mixture is essentially a two-component ceramic system. It is therefore of interest to examine the effects of these processing parameters on the property changes of the glass–mica system, particularly for the two distinctive mixtures which produced profoundly different physical structures and properties. These are the mixtures which consisted of 90 wt% glass and 10 wt% mica which produced a cellular structure and 80 wt% glass and 20 wt% mica which produced a highly densified ceramic body structure. Some of the more interesting experimental resulst are presented in this paper.

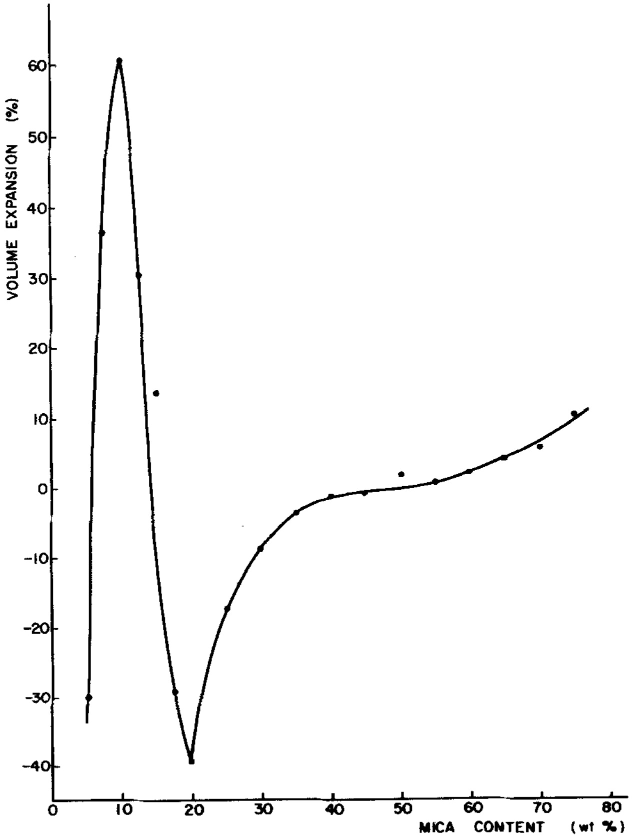

Figure 1 Variation of volume expansion with phlogopite mica content in the sintered composite compacts of recycled colourless soda–lime glass and phlogopite mica powders.

TABLE I Chemical analysis of the recycled colourless glass powders

Components	Weight %
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

TABLE II Chemical analysis of the phlogopite type mica powder

Components	Weight %
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

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 in diameter) by a mechanical pulverizer. The pulverized glass grains were then ground into fine powders by a dry ball-mill grinding process for a period of about 5 h. Corundum grinding media were used. The glass powders used for sample preparation were mostly in the 150 μm to 300 μm range. Chemical analysis showed that the recycled colourless glass powders consisted of the compositions shown in Table I.

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 shown in Table II [5].

For most experimental evaluation purposes, a 50 g specimen was prepared from the 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 before and after the addition of a wetting agent. The mixed powders were then compressed into a circular disc measuring about 50 mm in diameter and 20 mm in thickness. The compressed powder compact was then placed on a refractory brick and sintered in a Glo-Bar electric heating furnace under atmospheric conditions. 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 about 45 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 specimens from the furnace took about 6 h.

A wide range of powder compaction pressures, sintering temperatures and amounts of water which were added to the powder mixtures as wetting agent have been employed in the preparation of samples. This included the application of compaction pressures ranging from 1.4 to 17.5 MPa at several intervals, sintering temperatures varying from 780 to 900° C at 25° C intervals and amounts of water varying from 1 to 12 ml for a 50 g sample.

The physical properties of the sintered glass–mica composite samples were characterized by determining the bulk density, apparent porosity, water absorption and volume changes of each of the prepared samples according to the procedures described in the ASTM-C20 methods [6]. The compressive strengths of the sintered composite samples were measured with an INSTRON Universal Testing Instrument. The thermal conductivities of the prepared samples were determined with a Dynatech Guarded Hot-Plate according to the testing procedure described in the ASTM-C177 method [7] based on a larger sample measuring 20.0 cm in diameter.

3. Results and discussion

3.1. Effect of compaction pressures

It is generally known that the degree of densification of ceramic materials in a two-component

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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 Société Minéralurgique Laviolette, Inc. Montreal, under a trade name known as "SUZORITE" mica.

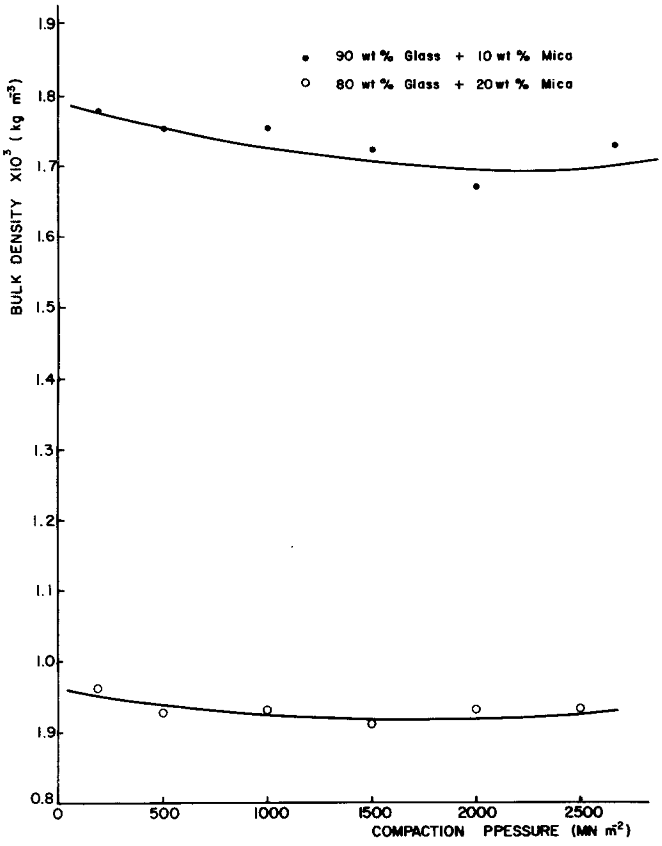

Figure 2 Effects of compaction pressures on the change of bulk density of the sintered composite compacts of recycled colourless soda-lime glass and phlogopite mica powders.

system is often influenced by the packing of the component particles in the compacted solid of the mixture. This is because it can affect the mechanism of material transfer in the densification process and because the packing of the particles is dependent on the applied compaction pressure [8]. The glass–mica mixture is essentially a two-component system; however, the physical appearance of the two component particles is significantly different. The ground glass powders are fine aggregates of irregular shape; while the mica powders are fine, thin flakes. The degree of particle packing resulting from the application of the various compaction pressures could therefore be an important processing parameter. In the present study, compaction pressures varying from 1.4 to 17.4 MN m^{-2} have been employed to compress the loose powders into compacted solids for sintering. However, experimental results showed that the powder compaction pressures have virtually no influence on the changes of the physical properties such as bulk density, volume expansion and apparent porosity of the glass–mica composite material for both series of samples prepared from the two specific compositions. All samples prepared from the mixture with 90 wt% glass and 10 wt% mica showed a cellular-structure, a bulk density value in the range of 0.915 to 0.962 × 10^3 kg m^{-3}, an average porosity in the range of 50 to 55% and an average volume expansion of about 40%. On the other hand, all samples prepared from the mixture with 80 wt% glass and 20 wt% mica showed a ceramic body of densified structure, a bulk density value in the range of 1.778 to 1.873 × 10^3 kg m^{-3}, an average volume contraction of about 32% and an apparent porosity in the range of 10 to 18%. As an example illustrating the non-sensitivity of the compaction pressure on the physical property change, Fig. 2 shows the variation of the bulk density as a function of the compaction pressures for samples prepared by sintering at 850° C for 30 min under atmospheric conditions. It is evident that the bulk density of the sintered composite samples is not affected by the compaction pressure. This could possibly be explained by the fact that the sintering temperature employed is above the melting point of the glass particles and that the mechanism of densification of the composite matrix is no longer governed by the degree of particle packing and

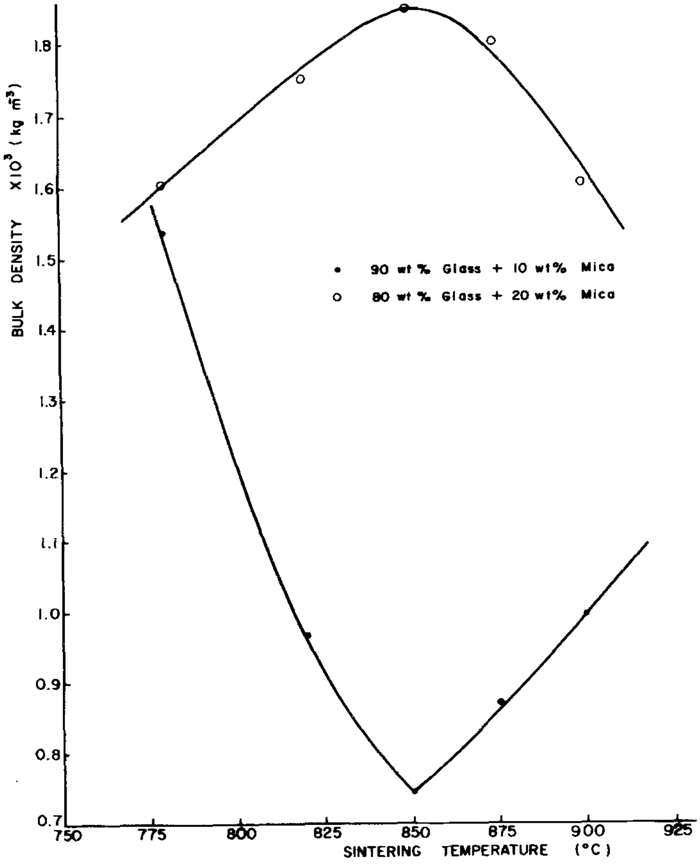

Figure 3 Effects of sintering temperatures on the change of bulk density of the sintered composite compacts of recycled colourless soda-lime glass and phlogopite mica powders.

subsequent material transfer, but is determined by the fluidity of the viscous liquid glass and the uniformity of the dispersion of the mica particles in the liquid phase glass in the matrix.

3.2. Effect of sintering temperatures

Temperature plays a significant role in the densification of ceramic materials by the solid-state sintering process. In the present study, sintering temperatures varying from 780 to 900° C have been employed to sinter the compacted glass–mica mixtures. Experimental results showed that the magnitude of the bulk density, the degree of volume expansion or contraction and the percentage of porosity present in the sintered bulk were all affected by the sintering temperature. Fig. 3 shows the variation of the bulk density of the glass–mica composite material with sintering temperatures for both compositions. It is evident that a very pronounced and opposite effect was observed in the two series of sintered samples. For mixtures composed of 90 wt% glass and 10 wt% mica, which produced the cellular-structure composite material, a minimum value of the bulk density was attained at 850° C. On the other hand, for mixtures composed of 80 wt% glass and 20 wt% mica, a reverse trend was observed with an optimum bulk density occurring at 850° C.

Under normal solid-state sintering processes, physical changes in the bulk of the compacted solid of ceramic materials that occur during the firing process are related to changes in grain size and shape, changes in pore shapes, or changes in pore size [8]. The porosity of the sintered glass–mica composite materials was found to vary with opposite trends between the two compositions. Fig. 4 shows the variation of the apparent porosity as a function of sintering temperatures. It is evident that the porosity of the sintered composite samples with the cellular-structure increased as the sintering temperatures increased; while the porosity of the sintered composite samples with the densified ceramic structure decreased as the sintering temperatures increased.

The display of opposite variations of the bulk density (or volume changes) and porosity of the two composite materials with sintering tempera-

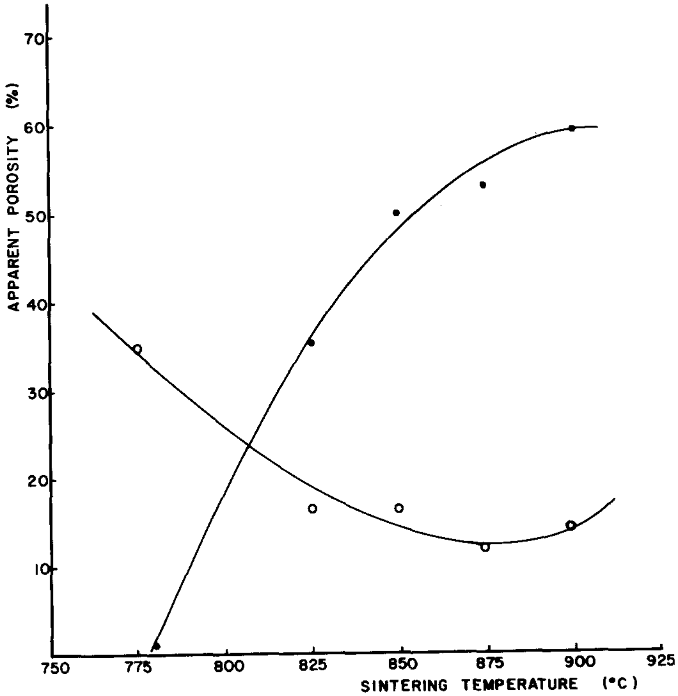

Figure 4 Effects of sintering temperatures on the formation of apparent porosity of the sintered composite compacts of recycled colourless soda-lime glass and phlogopite mica powders.

tures is probably attributed to the changes in the viscosity of the liquid phase glass in the composite matrix, that is caused by two contributing sources. As reported by the manufacturer [5], the phlogopite-type mica powders are known to contain about 0.01 wt% free water and 1.01 wt% combined water in the structure. Both free and combined water are known to be removable by elevated temperature heat treatment through an evaporation and dehydration process. An effective dehydration process begins at 500° C and is completed at approximately 1000° C. At the 800 to 900° C temperature level, it is estimated that about 60 to 70% of the combined water will be released from the mica particles by the dehydration process. The sintering temperature therefore determines the exact amount of water produced by the dehydration process. As the water is released from the mica particles in the matrix of the mixture, it is expected that a small portion of the water will be dissolved in the liquid phase glass. The dissolution of the water in the liquid phase glass will consequently cause a lowering of the viscosity of the glass–mica system. This was observed in the reaction of water with glass by other workers [9, 10]. Temperature also contributes to the lowering of the viscosity of the liquid phase glass [11]. The combined effects of these two contributing factors therefore significantly modified the viscosity of the glass–mica composite matrix and consequently determined the formation and arrangements of the glass network and the mica particle dispersion in the liquid phase glass.

Since only a small portion of the water produced by the dehydration process of the mica particles is dissolved in the liquid phase glass, the undissolved water will then be trapped in the interior of the glass–mica matrix in a vapour phase. The effectiveness of entrapping this water vapour depends on the closeness of the glass network surrounding the mica particles. The development of a close glass network around the mica particles is in turn dependent on the amount of the liquid phase glass present in the matrix. For a composition containing a large proportion of glass and a small proportion of mica, such as the composition consisting of 90 wt% glass and 10 wt% mica, a tighter glass network is produced. This will therefore be more effective in preventing the water vapour from escaping from the interior thereby allowing the water vapour to develop a high vapour pressure. This consequently caused the matrix to expand and caused the formation of a larger volume or decrease in bulk density. On the other hand, as the proportion of mica particles in the mixture is increased, the spreading of the liquid glass and the formation of a close glass network around the large number of mica particles are reduced. As a result, a greater fraction of the water vapour is allowed to escape from the matrix thereby weakening its expanding force. The absence of a strong expanding force therefore allows a more uniform dispersion and closer packing of the mica particles in the matrix during the solidification process. This results in the formation of a smaller volume of matrix, an increase in the bulk density and also of a lower porosity.

The occurence of an optimum bulk density or volume changes in the glass–mica composite material for both the cellular-structure and the densified-structure at 850° C is intuitively postulated due to the attainment of an equilibrium condition between the viscosity change resulting from the dissolution of water produced by the dehydration of the mica particles and the entrapment of the water vapour developed by the undissolved water at that critical temperature. Under the equilibrium condition the formation of a close glass network and a homogeneous dispersion of the mica particles in the matrix are balanced thereby allowing the establishment of a well co-ordinated structure during the solidification process.

3.3. Effect of water

Compaction of dry powders of ceramic materials for the solid-state sintering reaction is often facilitated by the addition of a bonding or wetting agent to the particle components. This is to improve the surface adhesion property of the particles. In the present study, a small quantity of water was added to the glass–mica powder mixtures prior to the compaction process to serve as a wetting agent. Experimental results showed that the quantity of water added to the dry mixtures of the glass–mica system has produced significant changes in the physical properties of the sintered glass–mica composite material. The changes in physical properties were particularly pronounced for the composite material having the cellular structure. Fig. 5 shows the variation of volume expansion as a function of the quantity of water added to a mixture of 50 g sample for both compositions. It is evident that the amount of the water added to the mixture consisting of 80 wt% glass and 20 wt% mica has practically no influence on the volume changes or the bulk density of the highly densified ceramic bodies. However, when a similar quantity of water was added to the mixture consisting of 90 wt% glass and 10 wt% mica, significant changes of volume expansion in the cellular structure were observed. Measurements show that the volume expansion of the cellular structure increased slightly as the quantity of water added to the mixture increased from 1 to 3 ml and a maximum volume expansion of greater than 60% was attained when 3 ml water was used. It must be pointed out that extreme difficulties were encountered in compressing a batch of mixtures consisting of both dry glass powders and mica powders without the presence of water as a wetting agent because of the lack of particle adhesion. It was found necessary to add at least 1 ml of water to a 50 g sample in order to overcome the compaction difficulty of the dry powders. When more than 3 ml of water was added to the mixture, the powder compaction process was considerably facilitated, but the volume expansion of the cellular-structure decreased non-linearly with the increase in quantity of water beyond the 3 ml level, as shown in Fig. 5.

The quantity of water added to the glass–mica mixtures as wetting agent during the powder compaction process affected not only the volume expansion, but also affected the apparent porosity of the sintered bulk. As shown in Fig. 6, sintered glass–mica composite samples of both structures exhibited the largest degree of porosity when either a minimum or an excess of water was added to the dry powder mixture in the compaction process. A minimum porosity was attained when about 5 to 7 ml of water which is equivalent to about 10% by weight of the powder sample was used. The presence of greater porosity in the sintered glass–mica composite samples when only a minimum quantity of water was added to the mixture is probably due to the formation of a large number of interconnecting pores which were developed because of the more viscous nature of the liquid glass. On the other hand, the development of greater porosity when excessive water is used may probably be attributed to the decrease in the viscosity caused by the large dissolution of water in the liquid phase and the poorer stability of the closed pores. The control of the quantity of water added to the glass–mica mixtures as wetting agent is therefore an important controlling parameter in the fabrication process of glass–mica composite material.

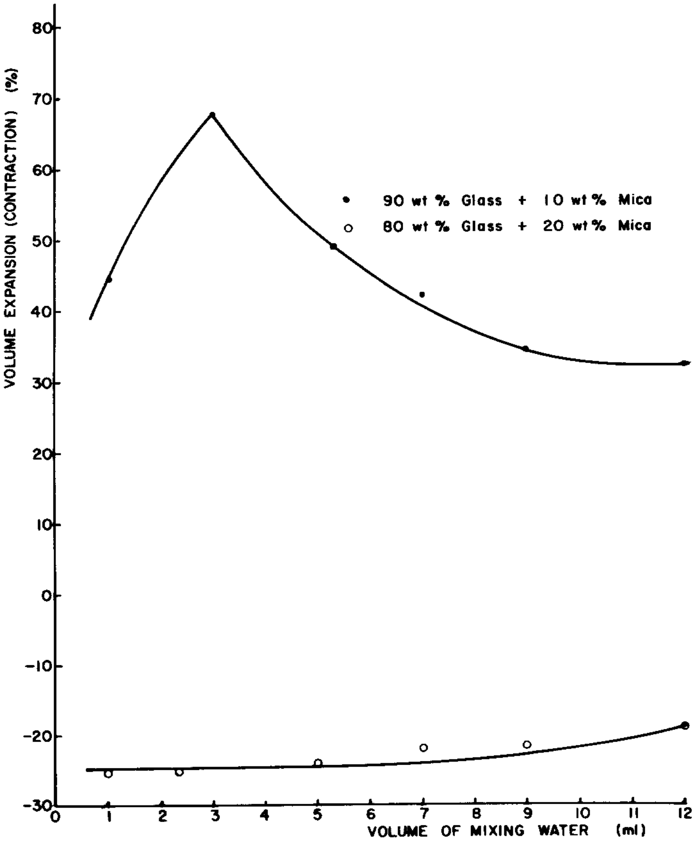

Figure 5 Effects of added water on the volume expansion (or contraction) of sintered composite compact of recycled colourless soda-lime glass and phlogopite mica powders.

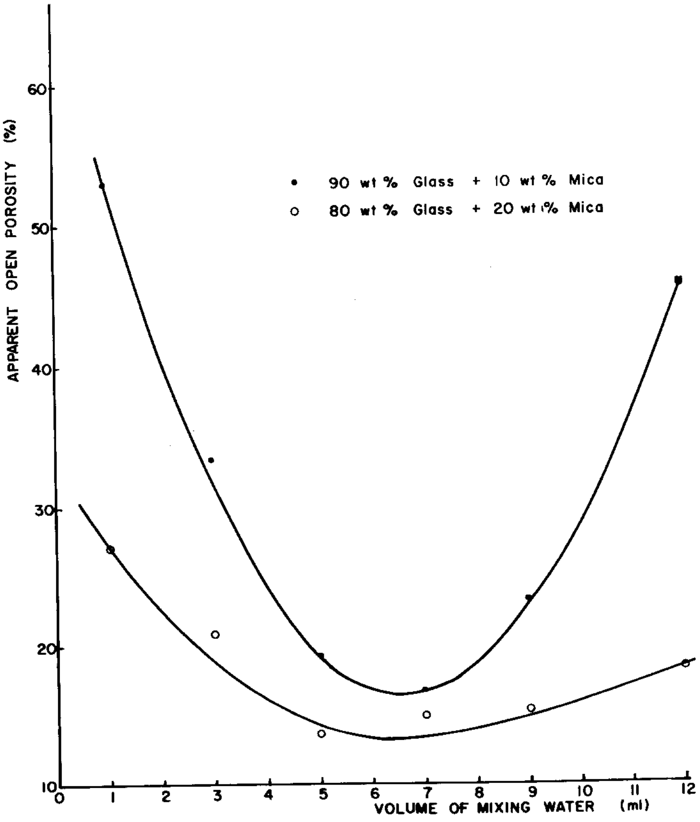

Figure 6 Effects of added water on the formation of porosity in the sintered composite compacts of recycled colourless soda-lime glass and phlogopite mica powders.

3.4. Mechanical and thermal properties

The effects of the various processing parameters on the physical properties of the glass–mica composite material have been systematic and conclusive. However, the effects of these processing parameters on the mechanical and thermal properties are less obvious and systematic.

Mechanical testing showed that both the powder compaction pressure and the quantity of water added to the powder mixtures as wetting agent have no significant influence on the compressive strength of the glass–mica composite material. Sintered samples with the cellular structure exhibited a compressive strength mostly in the range of 0.7 to 1.0 MN m^{-2}, whereas sintered samples with the highly densified ceramic structure exhibited a compressive strength varying from 42 to 59 MN m^{-2}. No trends of variation of the compressive strength with either the compaction pressure or the water content added to the mixture can be correlated. This can be seen from the data shown in Table III. As discussed, glass–mica composite material, when sintered at a temperature of 850° C exhibited a minimum bulk density for samples with the cellular structure and a maximum bulk density for samples with the highly densified ceramic structure. A similar trend was also observed when the compressive strength and the sintering temperature of the glass–mica composite samples were correlated. Table III shows the results for samples with the densified ceramic structure. The close relationship between compressive strength and the bulk density of the glass–mica composite material was found to be in agreement with the results obtained in a previous study [1].

The thermal conductivity of the glass–mica composite material with the cellular-structure as determined by the guarded hot-plate system was found to be in the range of 0.170 to 0.205 W m^{-1} °C^{-1} when measured in the temperature range 25 to 40° C and in the range 0.185 to 0.230 W m^{-1} °C^{-1} when measured in the temperature range 150 to 180° C. On the other hand, the thermal conductivity of the glass–mica composite material with the densified-structure was found to be in the range of 0.190 to 0.290 W m^{-1} °C^{-1} when measured in the temperature range 25 to 40° C and in the range of 0.205 to 0.310 W m^{-1} °C^{-1} when measured in the temperature range of 150 to 180° C. No obvious systematic correlation between the thermal conductivity of the sintered examples and the physical properties could be established. However, the overall thermal conductivity values of the various measured samples of the glass–mica composite material of both structures as measured at different temperature levels can be generalized and graphically plotted as shown in Fig. 7. It is evident that the thermal conductivity values of the sintered glass–mica composite material lie between the low values of the conventional insulating materials [12] and the high values of the oven-dried normal weight concrete and soda–

TABLE III Compressive strength of 80 wt% glass + 20 wt% mica composite materials prepared under controlled sintering temperature, compaction pressures and water contents as wetting agents

Sample number	Sintering temp. (° C)	Compaction pressure (MPa)	Water content (ml)	Compressive strength (MN m^{-2})
MC-2	780	1.37	3	21.55
MC-45	825	1.37	3	41.50
MC-19	850	1.37	3	53.03
MC-9	875	1.37	3	47.84
MC-42	900	1.37	3	11.40
MC-48	850	1.37	3	52.05
MC-49	850	3.44	3	56.30
MC-50	850	6.89	3	55.28
MC-51	850	10.34	3	41.83
MC-52	850	13.79	3	46.32
MC-53	850	17.24	3	47.29
MC-62	850	1.37	1	57.35
MC-63	850	1.37	3	52.97
MC-64	850	1.37	5	54.10
MC-65	850	1.37	7	59.86
MC-66	850	1.37	9	47.83
MC-67	850	1.37	12	50.35

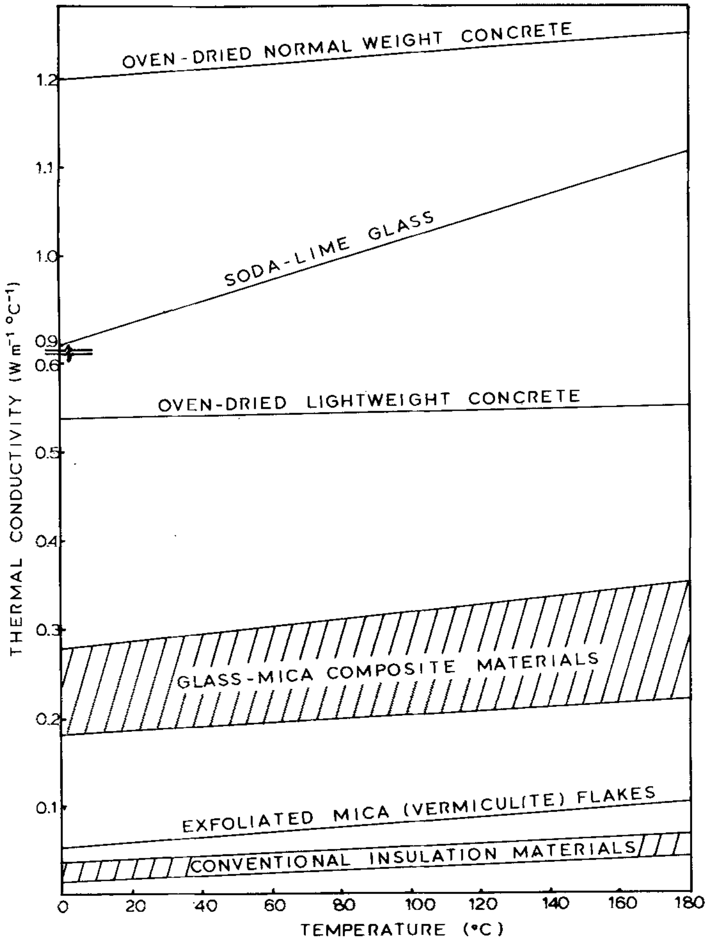

Figure 7 Thermal conductivity values of various conventional insulating materials, normal weight concrete, light-weight concrete, soda-lime glass and glass-mica composite materials as measured at different temperature levels.

lime glass [13, 14]. It also shows that the sintered glass–mica composite material has a much superior thermal insulating value compared with the insulating lightweight concrete.

4. Conclusions

The present study has demonstrated that the preparation of glass–mica composite materials with optimum physical, mechanical and thermal properties can be achieved by close control of the preparation parameters, such as sintering temperature and the quantity of water which is to be added to the mixture as wetting agent. Experimental results have shown that both the sintering temperature and the amount of water added to the mixture can affect several physical properties such as volume expansion, bulk density and apparent porosity. Experimental evidence obtained in the present study together with that reported in the previous paper [1] confirm that the two specific compositions chosen for the present study, i.e. 90 wt% glass + 10 wt% mica and 80 wt% glass + 20 wt% mica, are the unique compositions in the soda–lime glass and phlogopite mica system; as the former composition consistently produced a cellular structure composite material, while the latter composition consistently produced a highly densified ceramic structure.

This work has proved that glass–mica composite material with either the cellular structure or the densified ceramic structure can be prepared by the solid state sintering process at any temperature in the range 780 to 900° C. However, samples sintered at 850° C were found to exhibit optimum properties. Samples with the cellular structure showed maximum volume expansion thus resulting with a minimum bulk density when sintered at 850° C. Samples with the densified ceramic structure showed maximum densification and highest compressive strength when sintered at a similar temperature. There are indication that the sintering temperature 850° C is a critical temperature in the formation of the glass–mica composite material as an equilibrium state between the formation of glass network and the dispersion of mica particles in the liquid glass.

The quantity of water which is added to the glass–mica powder mixture as wetting agent was found to serve as more than just a wetting agent in facilitating the powder compaction process. Experimental results have shown that it can be used to control the percentage of the apparent porosity in the cellular structure of the glass–mica composite material. The addition of a water content equivalent to about 10% by weight was found to produce the lowest porosity in the glass–mica composite bulk.

Compaction pressure which is applied to compress the powder mixture of the two starting components into compacted solids for solid-state sintering was found to have no influence on either the physical or the mechanical properties. There are indications that property modification of the glass–mica composite material is controlled mainly by the uniformity of the melting of the glass powders and the dispersion of the solid mica particles in the liquid glass melt rather by the inter-particle contact or particle growth between neighbours.

The compressive strength of the glass–mica composite material with the densified ceramic structure is found to be superior than the compressive strength of several conventional building materials, such as building bricks (common brick) with compressive strength values in the range 17 to 55 $\mathrm{MN\,m^{-2}}$, non-reinforced concrete with compressive strength in the range 10 to 45 $\mathrm{MN\,m^{-2}}$ and cement mortar with compressive strength in the range 10 to 20 $\mathrm{MN\,m^{-2}}$. The thermal insulating value of the glass–mica composite material with either the cellular structure or the densified structure is definitely much superior than the concrete and masonry products. The display of both superior mechanical and thermal insulting properties of the glass–mica composite material suggests that the glass–mica composite material is a potential structural element for building construction applications as it may contribute to energy conservation.

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