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## Boron Promoted on Crystallization and Microstructure of Strontium Containing Mica Glass-Ceramics

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### ABSTRACT

The process of nucleation and crystallization of  $\text{SrO}\cdot 4\text{MgO}\cdot \text{Al}_2\text{O}_3\cdot 6\text{SiO}_2\cdot 2\text{MgF}_2$  glasses with  $\text{B}_2\text{O}_3$  as nucleating agent were characterized by differential thermal analysis, X-ray diffraction and scanning electron microscopy. A number of glass-ceramics of each glass batch with excess  $\text{B}_2\text{O}_3$  [SR2 (2 %  $\text{B}_2\text{O}_3$ ), SR4 (4 %  $\text{B}_2\text{O}_3$ ), SR6 (6 %  $\text{B}_2\text{O}_3$ ) and SR8 (8 %  $\text{B}_2\text{O}_3$ )] were made by heating at its respective nucleation temperature followed by different ceramization temperatures (800–1000 °C). The analysis of DTA result indicated that the crystallization peak ( $T_p$ ) and glass transition ( $T_g$ ) temperatures decreased with increasing  $\text{B}_2\text{O}_3$  content. It was also found that the higher  $\text{B}_2\text{O}_3$  content, the higher the aspect ratio of fluorophlogopite crystal.

### KEYWORDS

Glass, Ceramics, Nucleation, Crystallization, Microstructure.

### INTRODUCTION

Fluormica glasses are the most important class of modern advanced materials for industrial applications because of their technological importance (*e.g.* use in flat panel display substrates, fiber glass and photochromic glass) and their wide range of structural and dynamic questions that remains unanswered [1]. Boron is unique as a network former in that it can readily change between three or four coordinated by oxygen as composition and temperature are changed [2]. The exchange between these two coordination sites may be the source of thermodynamic property changes, *i.e.* viscosity, melting point and glass transition temperature.

Trisilicic alkaline mica ( $\text{K}\cdot\text{Mg}_3\cdot\text{Al}\cdot\text{Si}_3\cdot\text{O}_{10}\cdot\text{F}_2$ ) is formed through crystallization of glasses based on the composition of  $\text{SiO}_2\cdot\text{Al}_2\text{O}_3\cdot\text{MgO}\cdot\text{MgF}_2\cdot\text{K}_2\text{O}$ . Such glass-ceramics are important class of materials as they are machinable to high ( $\pm 10 \mu\text{m}$ ) tolerances [3]. These materials composed a 'House of cards' microstructure of mica crystals lead a quality of machinability due to cleave easily along the interfaces between layers while being machined [4-6].

Potassium fluormica crystal phase has lower strength and fracture toughness compared with the precipitation of barium fluormica crystal phase [7]. Henry and Hill [8,9] studied the nucleation and crystallization behaviour of barium fluorphlo-

## EXPERIMENTAL

gopites with addition of alumina and lithia. Greene *et al.* [10] reported the substitution of barium by potassium based on the composition of  $(1-Z)\text{BaO}:\text{ZK}_2\text{O}:(6-X)\text{MgO}:\text{XMgF}_2:(3-Q)\text{Al}_2\text{O}_3:\text{QB}_2\text{O}_3:8\text{SiO}_2$  system (where  $Z = 0, 0.25, 0.5, 0.75$  and  $1.0$ ,  $X = 2, 2.5$  and  $3.0$  and  $Q = 0, 0.5$  and  $1.0$ ). The result revealed that increases in molar volume, coefficient of thermal expansion and decrease in fractional glass compactness, microhardness and glass transition temperature values.

Mallik *et al.* [11] investigated the crystallization kinetics as well as crystallization behaviour, microstructure and mechanical properties of barium-potassium mixed fluormica glass through partial and complete substitution of potassium by barium. The result implies that machinability and strength can be modified by gradual substitution of potassium by barium and also by the duration of heat treatment time schedule. The influence of  $\text{B}_2\text{O}_3$  content on the crystallization and microstructure behaviour of barium fluormica glass, the results suggest that the increase in  $\text{B}_2\text{O}_3$  content decreases both glass transition ( $T_g$ ) and crystallization peak temperatures ( $T_p$ ) [12]. Also, the addition of  $\text{B}_2\text{O}_3$  indicates to promote growth of fluormica crystals increases with increasing the volatility of the growth medium.

The influence of  $\text{Al}_2\text{O}_3$  content on the crystallization and microstructure behaviour of strontium fluormica glass [13], the result implies that the second peak crystallization temperature ( $T_p^2$ ) appeared on alumina content ( $x \geq 1.5$ ) and lower percentage of alumina content shown the glass transition ( $T_g$ ) and first crystallization peak ( $T_p^1$ ) temperature shift to lower temperature end. The DTA and XRD results also showed that the two different peak crystallization temperatures correspond to the formation of strontium fluorphlogopite and strontium aluminum silicate. Mallik *et al.* [14] investigated the crystallization, microstructure and mechanical behaviour of strontium fluorphlogopite glass-ceramics by varying the fluorine content. The result indicated that the crystallization peak ( $T_p$ ) and glass transition ( $T_g$ ) temperatures decreased with increasing fluorine content which also lowers down the activation energy (E). Hardness and fracture toughness values are higher for less fluorine containing glass-ceramics when they are treated isothermally.

Some research works [15-17] have claimed the influence of  $\text{B}_2\text{O}_3$  on formation and microstructure of fluorphlogopite mica glass-ceramics system but information does not provide a clear perception. It would be in the scientific and technological point of view to determine the role of  $\text{B}_2\text{O}_3$  additions on the formation and microstructure of strontium fluorphlogopite glass-ceramics. The present work aims to follow two objectives to determine the effect of addition of progressively increasing the small amount of  $\text{B}_2\text{O}_3$  on (i) nucleation, crystallization and microstructure of strontium mica glass-ceramics  $\text{SrO} \cdot 4\text{MgO} \cdot \text{Al}_2\text{O}_3 \cdot 6\text{SiO}_2 \cdot 2\text{MgF}_2$  and (ii) to determine the effect of  $\text{B}_2\text{O}_3$  addition on crystal sizes.

**Parent glass synthesis:** Analytical grade chemicals in powder form were used as starting materials in this study. The powder chemicals were from Merck Specialties Pvt. Ltd. India. The chemical compositions (in g) of the glass batches varying on the different % excess of  $\text{B}_2\text{O}_3$  are presented in Table-1. Alkali earth containing mica glass-ceramics have high strength with less machinability property compared with alkali containing mica glass-ceramics [4]. In this work, an effort has been made to substitute strontium by potassium for achieving better strength and good machinability. Addition of varying amounts of  $\text{B}_2\text{O}_3$  in different glass composition for lowering the melting temperature of the glass batches and to obtain the high aspect ratio of fluormica crystal.

The weighed batch materials, after properly mixing were melted in a Pt/Rh crucible in an electrically heated furnace at  $1400^\circ\text{C}$  for about 4-5 h with occasional stirring with a quartz rod in order to achieve the homogeneity of the molten glass. After the melting operation is over, the molten mass was poured into a preheated cast iron mold to make rectangular slabs of dimension  $(50 \times 25 \times 10)$  mm. Immediately after casting, the prepared rectangular slabs of glass samples were released from the mold and were quickly introduced into an annealing furnace operating at  $650^\circ\text{C}$  and soaked for 1 h followed by natural cooling to room temperature. After annealing, the block was cut into pieces to about 1-2 mm thickness with the help of a precision low speed cutting machine (Buehler). For nucleation, these pieces of samples were fired at  $695\text{-}710^\circ\text{C}$  for 2 h. After this operation, the samples heated to the corresponding different crystallization temperatures at a rate of  $2^\circ\text{C}/\text{min}$  while the samples were kept at the crystallization temperatures for 5 h.

**Differential thermal analysis (DTA):** Differential thermal analysis was carried out by Shimadzu DT40 thermal analyzer with  $\alpha$ -alumina powder as a reference material. Four different glasses were crushed and finally ground to  $\sim 75\ \mu\text{m}$  suitable for DTA analysis. Non-isothermal experiments were performed by heating  $\sim 17\ \text{mg}$  glass sample at a heating rate of  $10^\circ\text{C}/\text{min}$  in the temperature range from ambient to  $1000^\circ\text{C}$ .

**X-ray powder diffraction (XRD):** X-ray powder diffraction was carried out for crystallized samples at different crystallization temperatures. All crystallized samples were heat treated using a heating rate of  $10^\circ/\text{min}$ . to the nucleation temperature of  $695\text{-}710^\circ\text{C}$ , soaked for 2 h at this temperature and heated again at  $2^\circ\text{C}/\text{min}$  to the corresponding crystallization temperature for 5 h and then followed by natural cooling to room temperature. The crystallized glass samples were ground to  $\sim 75\ \mu\text{m}$ . X-ray diffraction experiments were performed using X-ray powder diffractometer (PW 1830, Panalytical) using Ni filtered  $\text{Cu-K}\alpha$ , X-radiation with scanning speed of  $2^\circ(2\theta)$  per minute. The diffraction pattern was recorded within Bragg's angle ranges

TABLE-1  
CHEMICAL COMPOSITIONS (g) OF GLASS BATCHES AS A FUNCTION OF % EXCESS  $\text{B}_2\text{O}_3$

Glass batch	$\text{SrCO}_3$	$\text{MgO}$	$\text{SiO}_2$	$\text{Al}_2\text{O}_3$	$\text{MgF}_2$	$\text{B}_2\text{O}_3$ (% excess)
SR2	17.49	19.50	42.32	12.21	14.93	2.00
SR4	17.49	19.50	42.32	12.21	14.93	4.00
SR6	17.49	19.50	42.32	12.21	14.93	6.00
SR8	17.49	19.50	42.32	12.21	14.93	8.00

$10^\circ < 2\theta < 70^\circ$ . The phases were identified by JCPDS numbers (ICDD - PDF2 data base).

**Scanning electron microscopy (SEM):** The microstructural development of the different crystallized samples was observed applying back scattered electron imaging (BEI) mode of the scanning electron microscope (Hitachi, S3400N, Japan). Before study, crystal surface of all the samples were polished following standard procedures and finally with diamond paste. The polished samples were etched chemically by HF solution for 30 s.

## RESULTS AND DISCUSSION

**Differential thermal analysis:** DTA curves for four different glass samples at a heating rate of  $10^\circ\text{C}/\text{min}$  are shown in Fig. 1. In SR2 glass batch, only one crystallization peak ( $T_p$ ) temperature is visible. But two crystallization peaks was observed in SR4, SR6 and SR8 glass batches. In addition, an endothermic peak was observed around  $1000^\circ\text{C}$  in all glass batches. The endothermic peak indicated that the sample tends to melt. It is clear that as  $\text{B}_2\text{O}_3$  content increases, glass transition ( $T_g$ ) and crystallization peak ( $T_p$ ) temperatures decreases in all batches.  $\text{B}_2\text{O}_3$  reduces the number of bridging bonds in silica-based network. This result leads to a decrease in viscosity and consequently, in higher mobilities for different ions and ionic complexes operative in crystallization process of glasses. This result indicated to blow up crystallization rates and lower crystallization peak temperature [18]. First crystallization peak indicated that the formation of strontium aluminium silicate in all glass batches but the second crystallization peak indicated that the formation of strontium fluorophlogopite in SR4, SR6 and SR8 glass batches, which helps to identify by XRD results.

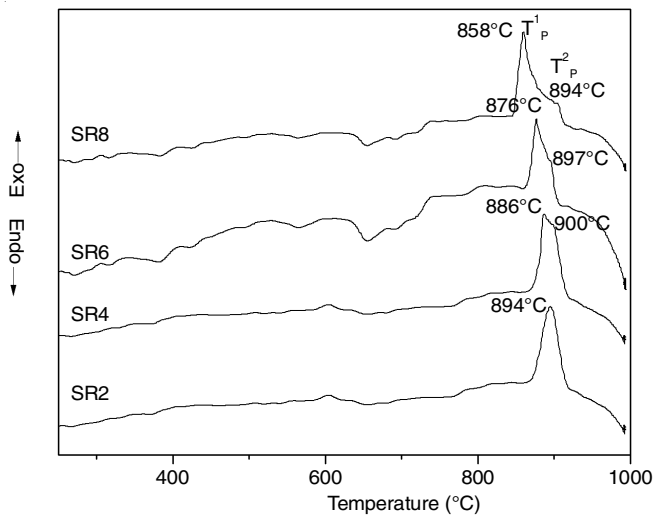


Fig. 1. Differential thermal analysis plots of glass samples with variation in  $\text{B}_2\text{O}_3$  content. The rate of heating is  $10^\circ\text{C}/\text{min}$ . (1) SR2 - 2 wt%  $\text{B}_2\text{O}_3$ , (2) SR4 - 4 wt%  $\text{B}_2\text{O}_3$ , (3) SR6 - 6 wt%  $\text{B}_2\text{O}_3$  and (4) SR8 - 8 wt%  $\text{B}_2\text{O}_3$

**X-ray diffraction:** X-ray diffraction patterns of crystallized samples of all four batches are shown in Figs. 2-5. Various crystal phases were identified by JCPDS reference files. Strontium fluorophlogopite is observed as a major crystal phases in all the heated samples of any particular batch. The amount of major phases increases with increasing ceramization temper-

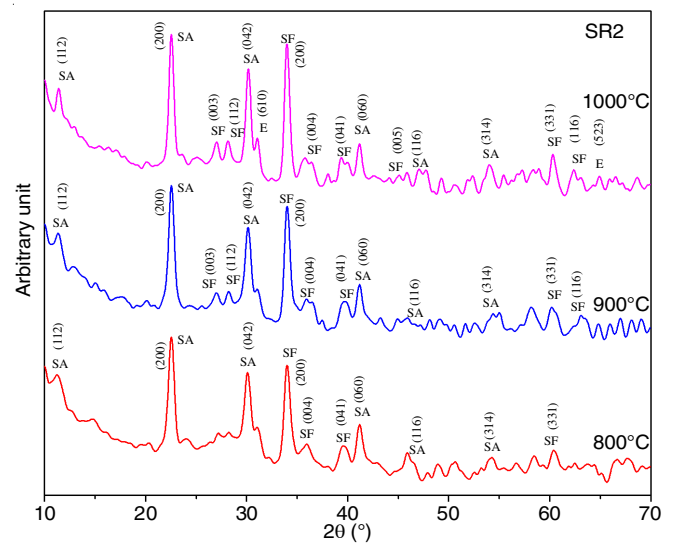


Fig. 2. XRD patterns for SR2 glass samples at different crystallization temperatures [SF-Strontium fluorophlogopite (JCPDS reference file - 019-0117), SA- Strontium aluminium silicate (JCPDS reference file - 01-088-1050) and E-Enstatite (JCPDS reference file - 019-0768)]

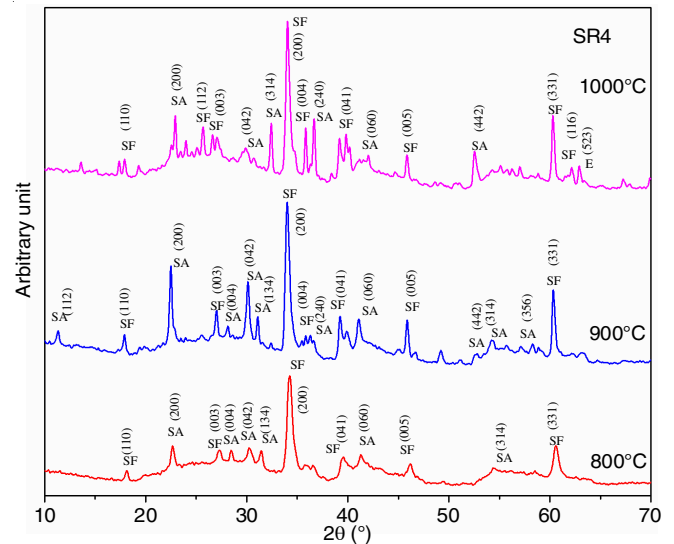


Fig. 3. XRD patterns for SR4 glass samples at different crystallization temperatures [SF-Strontium fluorophlogopite (JCPDS reference file - 019-0117), SA- Strontium aluminium silicate (JCPDS reference file - 01-088-1050) and E-Enstatite (JCPDS reference file - 019-0768)]

ature. Peaks of strontium aluminium silicate (SA) and enstatite (E) at  $800^\circ\text{C}$  correspond to a new phase other than major crystal phase of strontium fluorophlogopite (SF). From  $800$  to  $900^\circ\text{C}$ , there is no change in the intensity of the peaks and existing peaks remains unchanged. Two new crystal peaks of strontium fluorophlogopite appears at ( $2\theta = 18^\circ$ ) and SA ( $2\theta = 11.3^\circ$ ) in all the batches. The sharpness of the peaks of both strontium aluminium silicate and enstatite increases at  $900$  and  $1000^\circ\text{C}$ . It is noticed that with increase in crystallization temperature, the formation of strontium aluminium silicate decreases for any particular batch, but the formation of the strontium aluminium silicate seems to be major crystal phase in SR2 specimen heated at  $800$  and  $900^\circ\text{C}$  but not in SR4, SR6 and SR8 batches heated at  $800$  and  $900^\circ\text{C}$ . This may be an indication that strontium aluminium silicate formation takes place at lower temperature in all the batches.

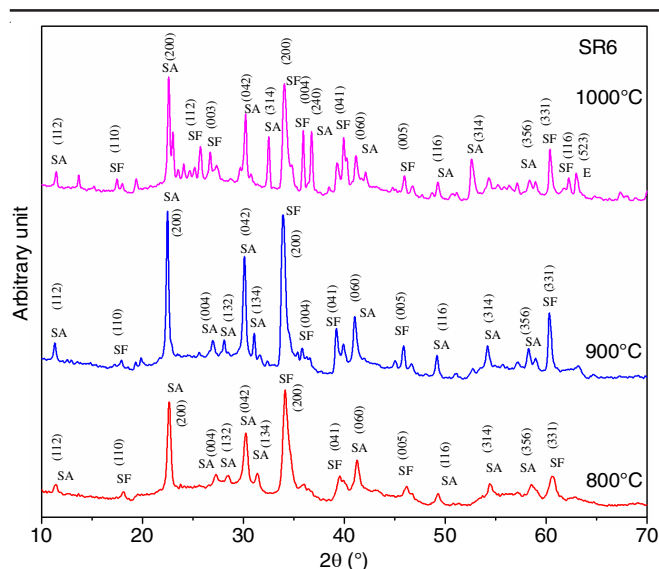


Fig. 4. XRD patterns for SR6 glass samples at different crystallization temperatures [SF-Strontium fluorphlogopite (JCPDS reference file - 019-0117), SA- Strontium aluminum silicate (JCPDS reference file - 01-088-1050) and E-Enstatite (JCPDS reference file - 019-0768)]

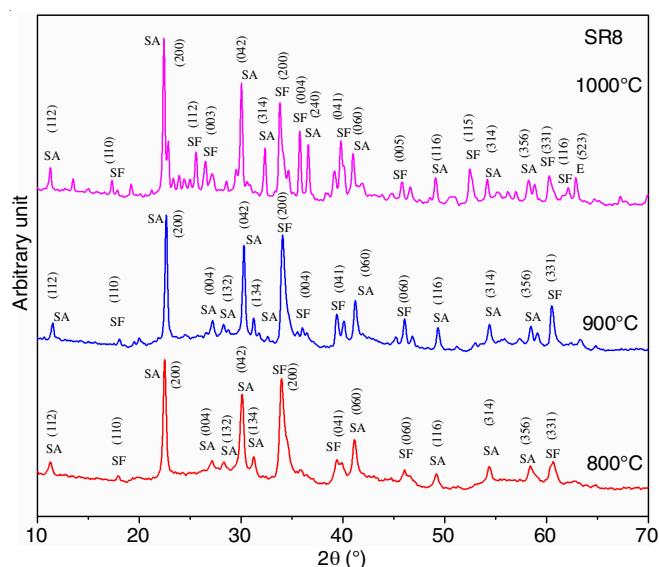


Fig. 5. XRD patterns for SR8 glass samples at different crystallization temperatures [SF-Strontium fluorphlogopite (JCPDS reference file - 019-0117), SA- Strontium aluminum silicate (JCPDS reference file - 01-088-1050) and E-Enstatite (JCPDS reference file - 019-0768)]

**Scanning electron microscope:** Microstructure development of all four different batches of glass sample crystallized at 800, 900 and 1000 °C for 5 h is shown in Figs. 6, 8-10.

At 800 °C, liquid-liquid phase separation occurs in SR2, SR4 and SR6 batch, respectively. The droplets are small and randomly distributed throughout the glass matrix (Fig. 6a, 8a and 9a). Sample crystallized at 900 °C exhibited a large number of slightly bigger block crystals (Fig. 6b, 8b, 9b and 10a). The large number of slightly bigger blocky crystal indicates that may lead to the formation of strontium aluminium silicate. This microstructure is commonly a high crystalline, blocky arrangement, very low aspect ratio [19]. The crystallization temperature increases up to 1000 °C (Fig. 6c) resulted in sufficient crystal growth and the crystal phases were characterized by EDX analysis (Fig. 7) as strontium fluorphlogopite.

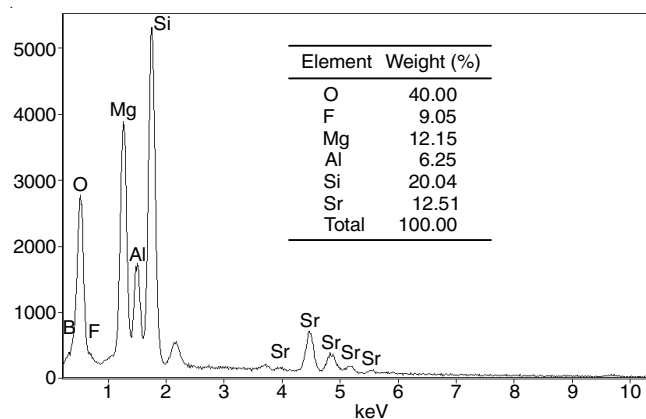


Fig. 7. EDX analysis of crystal phase of sample (SR2) crystallized at 1000 °C

Batches (SR4 and SR6) of sample heated at different crystallization temperature, undergo complex crystallization process that eventually leads to the formation of strontium fluorphlogopite crystals at 1000 °C in Figs. 8c and 9c. However, a large sized crystal appeared at 900 °C (Fig. 10b) in SR8 batch. After a 5 h heat treatment at 1000 °C, the mica crystals are observed in the microstructure with average crystal size of 4-5 μm (Figs. 6c, 8c and 9c). In SR8, large size of fluorphlogopite (13-16 μm) crystal appeared at heat treatment temperature 900 and 1000 °C (Figs. 10b and 10c).

The increasing B<sub>2</sub>O<sub>3</sub> content in above mentioned different glass batches consequently increases the rod-shaped size of the developed microstructure. These findings certify that B<sub>2</sub>O<sub>3</sub> promotes to display their natural tendency towards directional growth of rod-shaped crystals and controlled by slow diffusion rates.

**Comparative study:** Greene *et al.* [10] indicated that the melting temperature of barium-containing glass is 1450 °C. It

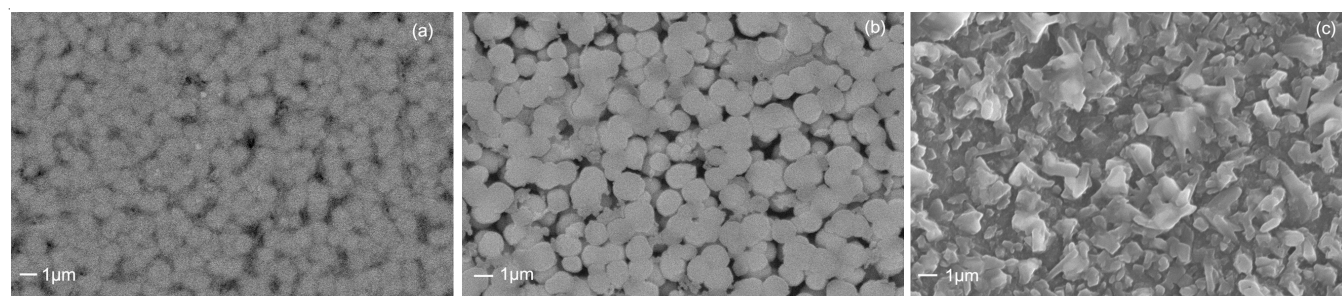


Fig. 6. SEM photograph of polished and etched surface of SR2 nucleated at 710 °C for 2 h and crystallized at 800 °C (a), 900 °C (b) and 1000 °C (c) for 5 h



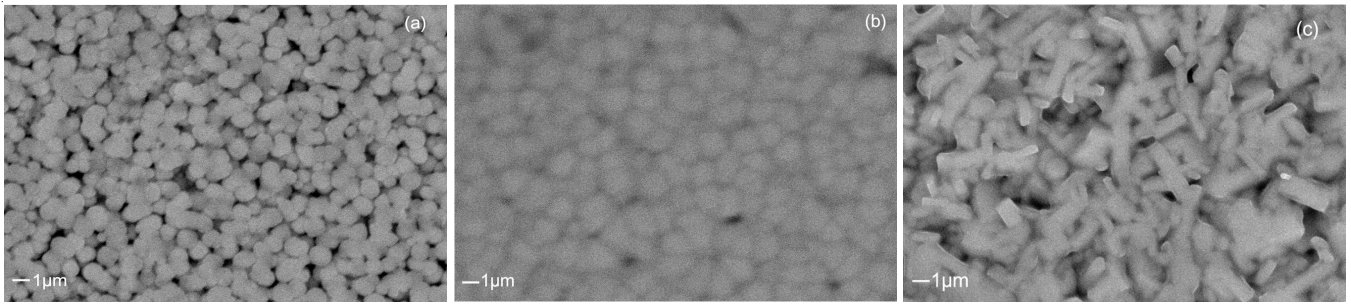


Fig. 8. SEM photograph of polished and etched surface of SR4 nucleated at 705 °C for 2 h and crystallized at 800 °C (a), 900 °C (b) and 1000 °C (c) for 5 h

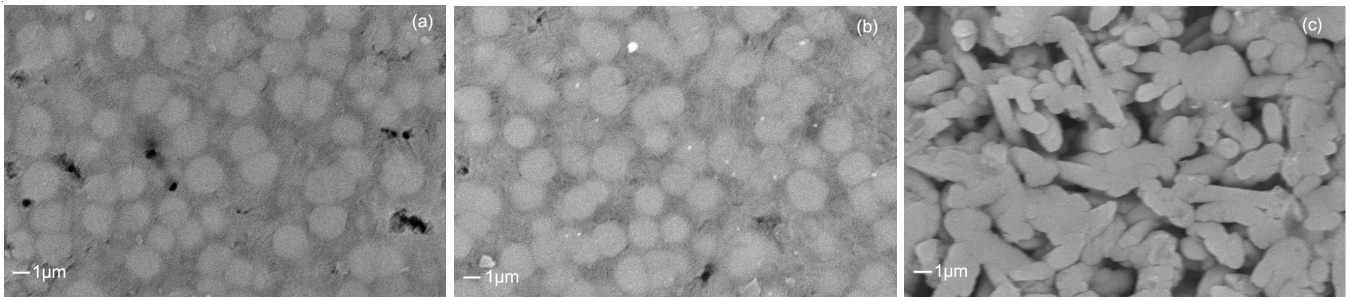


Fig. 9. SEM photograph of polished and etched surface of SR6 nucleated at 700 °C for 2 h and crystallized at 800 °C (a), 900 °C (b) and 1000 °C (c) for 5 h

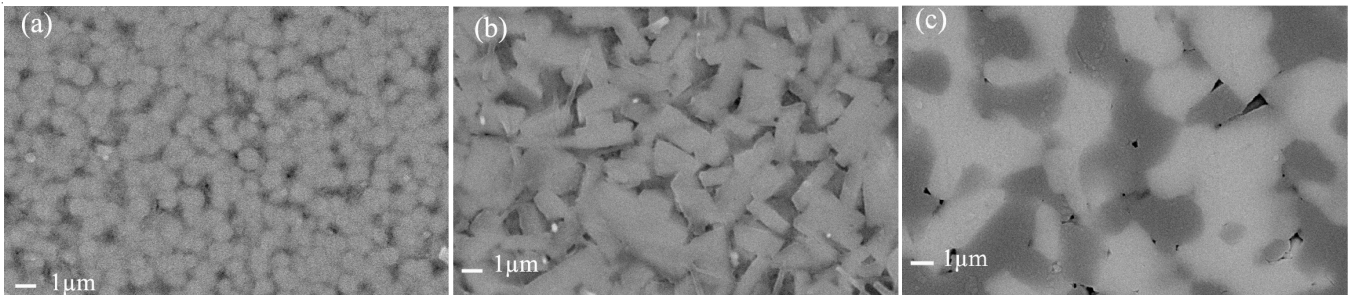


Fig. 10. SEM photograph of polished and etched surface of SR8 nucleated at 695 °C for 2 h and crystallized at 800 °C (a), 900 °C (b) and 1000 °C (c) for 5 h

also indicated that only one crystallization peak was observed, which is identified as barium fluorphlogopite in DTA analysis. X-ray diffraction analysis revealed that the formation of barium fluorphlogopite takes place at lower temperature. With increasing heat treatment temperature, the fluorphlogopite crystal size increases (12-15 μm in length) and also higher percentage of B<sub>2</sub>O<sub>3</sub> leads to the larger size of fluorphlogopite crystals, which is indicated in SEM results.

In comparison with present article, the results indicated that the melting temperature is 1400 °C. DTA analysis results revealed that the only one crystallization peak is observed in SR2 batch but two crystallization peaks were observed in SR4, SR6 and SR8 batches, respectively. First crystallization peak corresponds to the formation of strontium aluminum silicate in all batches and second crystallization peak corresponds to the formation of strontium fluorphlogopite, which is supported by XRD analysis. The XRD analyses results indicated that the formation of strontium aluminum silicate takes place at lower temperature. SEM analyses results also indicated that the formation of bigger blocky crystal of strontium aluminum silicate (4-5 μm in length) and the formation of larger size crystal of strontium fluorphlogopite (13-16 μm in length) [17].

## Conclusions

- DTA analyses results indicates that the glass transition ( $T_g$ ) and crystallization peak ( $T_p$ ) temperature shifted to lower temperature end for the samples becoming higher concentration of B<sub>2</sub>O<sub>3</sub> content. Appearance of crystallization peak temperature heralds to the formation of strontium fluorphlogopite.

- XRD analyses indicated that no other crystal phases appear prior to the formation of strontium fluorphlogopite, although the latter finally turns to enstatite and strontium aluminium silicate phases at 1000 °C.

- The addition of B<sub>2</sub>O<sub>3</sub> promotes the crystallization and grain growth of fluormica flakes by increasing the fluidity of the growth medium.

- Aspect ratio of the main crystals increased with increase in heat treatment temperature and with higher concentration of B<sub>2</sub>O<sub>3</sub> content in the composition of glass.

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