ARTICLE IN PRESS

Ceramics International xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Ceramics International

journal homepage: www.elsevier.com/locate/ceramint

Development of low thermal expansion mono crystalline Sr-feldspar phase via Sr-cordierite ceramic/borosilicate glass composite

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ARTICLE INFO

Keywords:
Composite
Sr-feldspar
Thermal expansion
Dielectric constant

ABSTRACT

The present work focuses on fabrication of low thermal expansion monoclinic Sr-feldspar through sintering of Sr-cordierite ceramic/borosilicate glass composite. The prepared composites were sintered between 1200 and 1350 °C. Bulk density and apparent porosity of sintered composites were measured according to Archimedes technique. Phase composition and microstructure of sintered composites were tested by X-ray diffraction and scanning electron microscope attached with EDAX unit. Thermal expansion coefficient and dielectric constant of sintered composites were also determined. The results revealed that mono crystalline Sr-feldspar was formed after sintering up to 1350 °C especially in the composites that contain higher ceramic content. As indicated from the result of microanalysis conducted by EDAX, the obtained Sr-feldspar was deficient in SiO_2 and SrO. In the composites with higher glass content, little amounts of quartz or cristobalite were also formed, depending on the cooling conditions. The sinterability was increased with increasing sintering temperature and decreased with increasing ceramic content. The obtained sintered composites exhibited low thermal expansion coefficient and dielectric constant. The composite that contains 90% ceramic exhibited thermal expansion coefficient 2.533×10^{-6} °C^{-1} and dielectric constant value 8.42.

1. Introduction

In the recent years, alkali and alkaline earth aluminum-silicate ceramics and glass-ceramics have paid a great of technological attention owing to their low thermal expansion, low dielectric constant and losses, good mechanical characteristics, and appropriate melting point. These characteristics make them important for many applications such as electronics, protecting coatings, thermal stable materials and biomedical applications [1–13]. Ceramic or glass-ceramic materials based on alkaline earth feldspare comprising calcium aluminum silicate ($CaAl_2Si_2O_8$), strontium aluminum silicate ($SrAl_2Si_2O_8$) and barium aluminum silicate ($BaAl_2Si_2O_8$) are getting special interest and used in different purposes. For $SrAl_2Si_2O_8$ compounds, there are three polymorphs crystallized in the forms of Sr-feldspar, Sr-paracelsian and Sr-hexacelsian. Sr-feldspar is the most stable form between 500 and 1650 °C, whereas Sr-paracelsian is stable from room temperature up to 500 °C. On the other hand, Sr-hexacelsian is metastable and can be formed after quenching the superheated melt. However both Sr-feldspar and Sr-hexacelsian are well known as synthetic product whereas Sr-paracelsian is known as natural product called slawsonite [14]. In $SrAl_2Si_4O_8$ glass system, the crystallization either in bulk or powder state needs low energy (451–534 kJ/mol) and gives fibrous microstructure, whereas $BaAl_2Si_2O_8$ glass requires higher energy (473–560 KJ/mol) for crystalization. In both glasses, hexacelsian is formed first then transforms into monocelsian at higher temperature [15]. Strontium-feldspar $SrAl_2Si_2O_8$, is a monoclinic framework structure which is closely identical to $BaAl_2Si_2O_8$ (Ba celsian) and may refer to Sr celsian which is stable up to 1700 °C [16]. Monoclinic Sr-feldspar glass-ceramic have good refractory property; at 1450 °C can resist deformation. It has low thermal expansion and dielectric properties as compared with hexacelsian. Beall, et. al developed Sr-feldspar in SrO-Al_2O_3-SiO_2 system with the presence of 8–16% TiO_2 [17]. Sr-Ba feldspar glass-ceramics have many interested applications in electronics as suitable substrates for semiconductors, particularly silicon. For the system $Ba_{1-x}Sr_xAl_2Si_2O_8$ (x = 0–1), the increase of strontium percent enhances the monoclinic phases and lowers the dielectric constant [18].

The goal of the present work is to prepare sintered low thermal expansion mono crystalline Sr-feldspar ceramic phase through the solid state reaction of nominal Sr-cordierite with borosilicate glass. The study focuses also on the sinterability, microstructure, physical, thermal and electrical properties of prepared materials.

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https://doi.org/10.1016/j.ceramint.2018.04.213
Received 11 April 2018; Received in revised form 23 April 2018; Accepted 24 April 2018
0272-8842/

Table 1
Chemical composition of Sr-cordierite ceramic and borosilicate glass as well as batch composition of designed composites.

Batch	Constituents						
	SiO_2	Al_2O_3	SrO_2	B_2O_3	Na_2O	Composite	
Glass	80.80	2.3	–	10.80	6.84		Borosilicate glass wt%
Ceramic	42.22	28.66	29.12	–	–	Sr-cordierite wt%	
GC5	61.51	15.48	14.75	5.4	3.42	50	50
GC6	57.65	18.12	17.47	4.32	2.74	60	40
GC7	53.79	20.75	20.38	3.24	2.05	70	30
GC8	49.94	23.39	23.30	2.16	1.37	80	20
GC9	46.08	26.02	26.21	1.08	0.68	90	10

Table 2
Bulk density and apparent porosity of sintered materials.

Property	Sintering temp., °C	Composite Samples				
		GC5	GC6	GC7	GC8	GC9
Density g/cm³	1200	2.283	2.064	2.008	1.955	1.960
	1350	2.60	2.55	2.21	2.00	2.04
Porosity %	1200	10.50	11.80	22.09	26.70	26.30
	1350	1.500	2.600	3.97	21.10	21.70

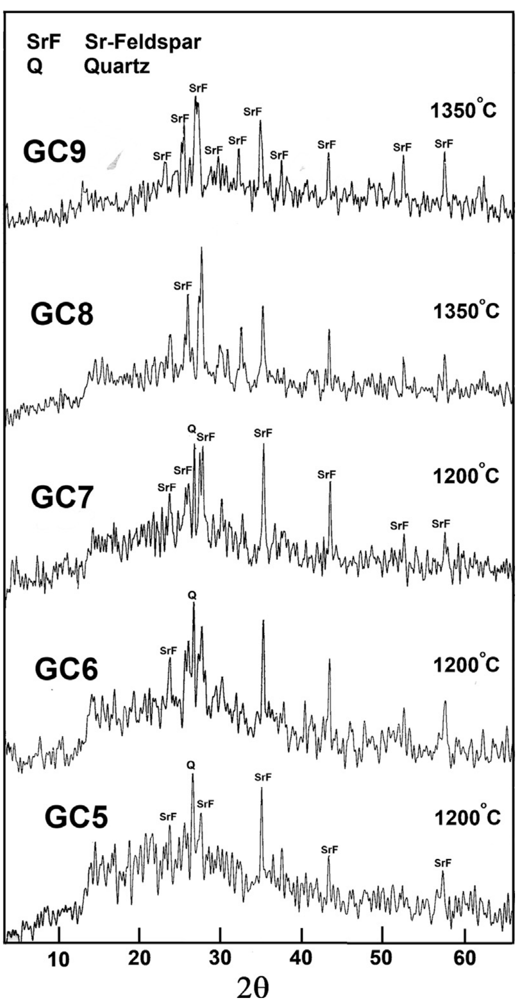

Fig. 1. XRD patterns of sintered composites normally cooled.

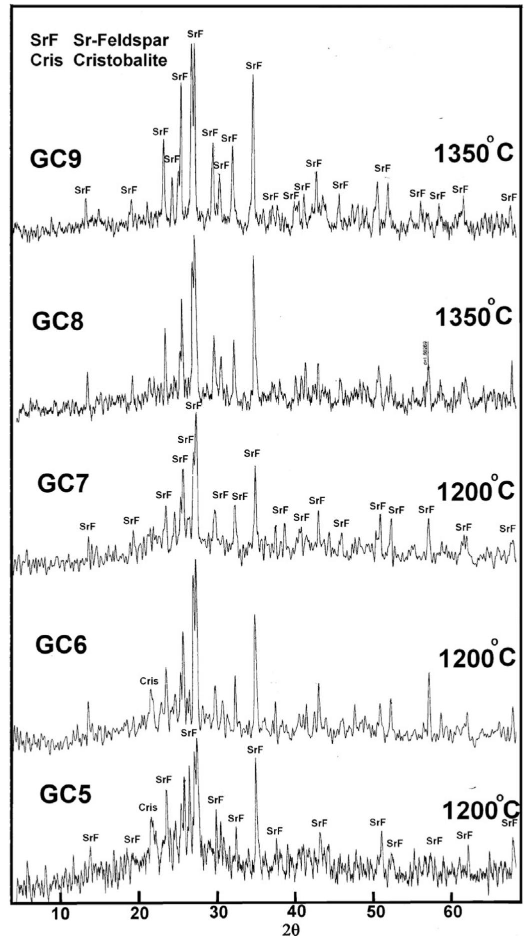

Fig. 2. XRD patterns of sintered composites fast air-quenched.

Table 3
Crystalline phases developed after sintering process.

Sample No	Sintering temperature	Crystalline phases	
		Normal cooling	Fast cooling
GC5	1200 °C /1 h	Low qz Sr-feldspar	Sr-feldspar Crist (low)
GC6	1200 °C /1 h	Low qz Sr-feldspar	Sr-feldspar Crist (low)
GC7	1200 °C /1 h	Low qz Sr-feldspar	Sr-feldspar
GC8	1350 °C /1 h	Sr-feldspar	Sr-feldspar
GC9	1350 °C /1 h	Sr-feldspar	Sr-feldspar

* Fast air quench after sintering process.

2. Materials and experimental methods

Highly pure chemicals as $SrCO_3$, Na_2CO_3, alpha Al_2O_3, H_3BO_3 and silica sand were supplied by Sigma-Aldrich and used in perpetration of borosilicate glass and Sr-cordierite ceramic ($Sr_2Al_4Si_5O_{18}$). Borosilicate glass within SiO_2–B_2O_3–Na_2O system was prepared in platinum crucible at 1500 °C as described elsewhere [13]. On the other side, stoichiometric Sr-cordierite was prepared from $SrCO_3$, alpha-Al_2O_3 and SiO_2 after well mixing and heating at 1200 °C. The chemical composition of prepared borosilicate glass and ceramic as well as designed batch compositions of desired composites are illustrated in Table 1. For preparation of Sr-feldspar, the glass and ceramics were well mixed according to the designed batch ratios using suitable amount of 7% PVA binder. The mixed powders were uniaxially pressed at 10 kN in a die of