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Sintering characteristics of in situ formed low expansion ceramics from a powder precursor in the form of hydroxy hydrogel

S. Mandal, S. Chakrabarti*, Sukhen Das, S. Ghatak

Central Glass & Ceramic Research Institute, Kolkata 700 032, India

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Abstract

Lithium aluminosilicate powder precursors of compositions Li_2O:Al_2O_3:SiO_2 as 1:1:2; 1:1:2.5 and 1:1:3 were prepared in the hydroxy hydrogel form by wet interaction technique in aqueous medium followed by sintering for ultimate synthesis of low expansion ceramics. Phases formed in the sintered specimens were analyzed by XRD technique. Thermal expansion of the specimens sintered at 1100, 1200 and 1300 °C were also measured. It was found that β-spodumene, lithium aluminum oxide and silica were the predominat phases in all the specimens. Sintering was optimum up to 1200 °C beyond which no further noticeable shrinkage was observed. The sintered specimens remained highly porous even after firing at 1300 °C, whose bulk density and apparent porosity were in the range of 1.25–1.42 g/cm^3 and 43–48%, respectively. Thermal expansion characteristics and density of the sintered specimens were found to be primarily related to the composition of the phases formed during sintering. A porous low expansion ceramic monolith could be prepared using the present technique.

Keywords: A. Sintering; Low expansion ceramics; β-Spodumene; Hydroxy hydrogel process

1. Introduction

The invention of lithium aluminosilicate glass–ceramics with extremely low thermal expansion provided a basis for producing materials for a broad range of use. The synthesis of crystalline species of low thermal expansion has long been an important facet of ceramic technology. Glass–ceramics based on the crystalline phases of the composition $Li_2O{\cdot}Al_2O_3{\cdot}nSiO_2$ are famous for ultra-low or even negative thermal expansivity over an extended temperature range. They have been widely used as cook-top panels, stove windows, and cookware and in precision parts [1].

In the system Li_2O–Al_2O_3–SiO_2, the mode of linking of the tetrahedral in the quartz structure is retained, but some Si atoms are replaced by Al while cations (Li^+, Mg^{2+}) are accommodated in some of the interstices to maintain the charge balance [2]. β-Quartz solution phases of this type have found tremendous application for low expansion glass ceramic [3]. The high temperature structure of β-eucryptite ($LiAlSiO_4$) can be imagined as a superstructure on β-quartz [4]. For high SiO_2 contents, however, such phases are only metastable precursors of the stable equilibrium phase (β-spodumene solid solution) [5]. This is confirmed by the study on the phase diagram in the system Li_2O–Al_2O_3–SiO_2 reported by Anon [6] in which the stability field of β-eucryptite ($LiAlSiO_4$) solid solution is confined to the range of compositions 1:1:1 to about 1:1:1.60 (Li:Al:Si). Mazza et al. [7] reported the high quartz solid solution phases by an unconventional sol–gel technique, by means of the preliminary formation of an amorphous solid (xerogel) with composition $Li_2O{\cdot}Al_2O_3{\cdot}nSiO_2$ ($n = 2$–4).

The present day method of making the material with low thermal expansion coefficient was reviewed in an earlier communication [8]. The phase relationships in the β-spodumene precursor powders have been examined earlier by using several methods [9–14]. Kobayashi et al. [9] have pointed out that the phases developed are β-spodumene and $LiAlSi_3O_8$ in the alumina sol, silica sol and $LiNO_3$ solution system. Suzuki et al. [10] have used the partial hydrolysis method of tetraethyl orthosilicate (TEOS) coupled with the double alkoxide method, resulting in copolymerization of the precursor solutions with a stoichometric β-spodumene composition and crystallization of powder precursors to the χ-spodumene (β-eucryptite/β-quartz solid solution) above

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* Corresponding author. Fax: +91-33-2473-0957/2483-8085.
E-mail address: chakrabarti_s@cgcri.res.in (S. Chakrabarti).

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Table 1
Compositional analysis after calcination at 750 °C

Constituents	Composition (wt.%) of the samples			Molar ratio
	S1	S2	S3	
Li_2O	10.54	9.17	8.71	S1 = LAS 1:1.01:2.52
Al_2O_3	36.21	34.37	30.18	S2 = LAS 1:1.10:3.04
SiO_2	53.25	56.46	61.11	S3 = LAS 1:1.02:3.51

LAS: lithium aluminosilicate.

Table 2
Physical properties of the sintered specimens sintered at different temperatures

Sample no.	1100 °C	1200 °C	1300 °C	1350 °C
(A) WA (%) at different sintering temperatures				
S1	36.71	32.07	32.45	32.64
S2	43.83	41.05	38.41	36.28
S3	32.39	31.17	31.87	32.12
(B) BD (g/cm^3) at different sintering temperatures				
S1	1.32	1.38	1.42	1.46
S2	1.18	1.21	1.25	1.28
S3	1.29	1.34	1.36	1.39
(C) AP (%) at different temperatures				
S1	47.12	42.86	43.45	44.04
S2	51.56	49.58	47.87	46.39
S3	44.26	43.07	44.51	44.97
(D) Fired shrinkage (%) at different temperatures				
S1	6.61	8.43	8.64	8.87
S2	4.04	4.68	5.88	6.12
S3	10.18	10.50	10.11	9.77

600 °C. The χ- and β-transformation of the spodumene occurs about 900 °C. Yang and co-workers [11] also have reported that on heating gel monoliths with a composition of β-spodumene, the β-eucryptite crystals are first precipitated around 750 °C, followed by precipitation of the β-spodumene crystals at 830 °C. Phalippou et al. [12] have reported that using $Si(OCH_3)_4$, $Al(NO_3)_3$ and $LiNO_3$ as the starting materials, the first crystallization phases observed by XRD are Li_2SiO_3 and β-spodumene. Veltri and Scola [13] have found that using $Si(OC_2H_5)_4$, $Al(OC_4H_9)_3$ and $LiNO_3$ as the starting materials and hot pressing at 1050 °C under 6.9 MPa for 15 min, the crystallized phase observed by X-ray diffraction (XRD) are β-spodumene and mullite. Using $Si(OC_2H_5)_4$, $Al(OC_2H_5)_3$, and $LiOCH_3$ as the starting materials and heating the resulted gels from 400 to 1200 °C, Wang [14] has shown the diffraction of β-spodumene. Wang et al. [15] reported that β-spodumene ($LiAl_2O_6$) has a tetragonal dipyramid crystal structure which is uniaxial positive. Li and Peacer [16] pointed out that the structure of β-spodumene consists of a three-dimensional network Si–O and Al–O tetrahedral. These tetrahedral are randomly distributed in the network; with the Li ions in the interstitial positions. It was reported later on [17–19]

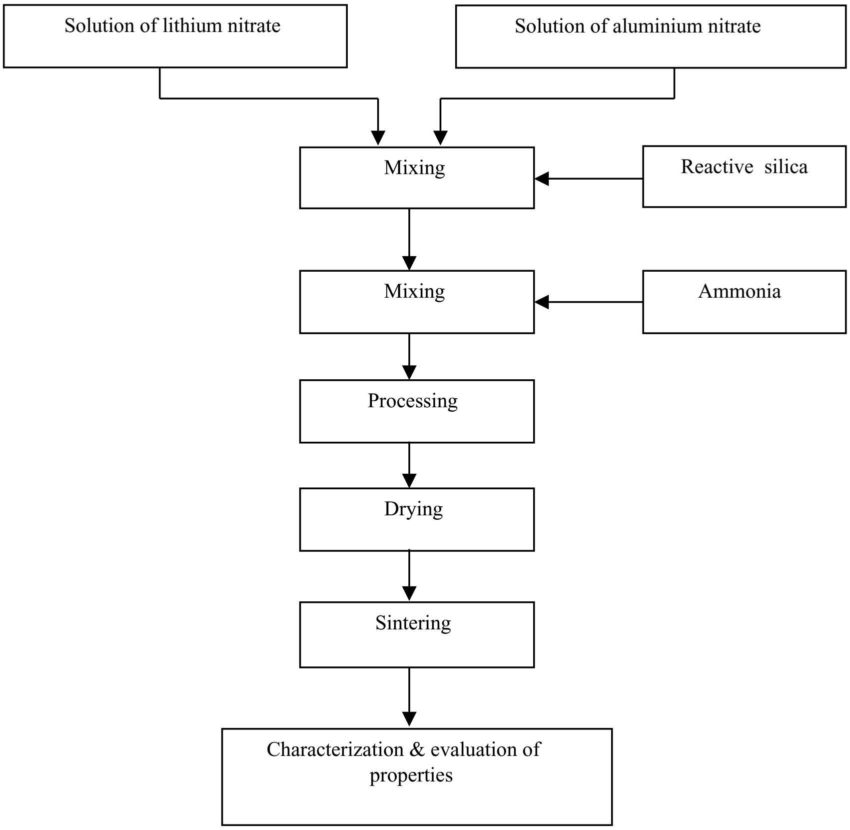

Fig. 1. Process for preparing porous low expansion ceramic monolith from “hydroxy hydrogel” powder precursor.

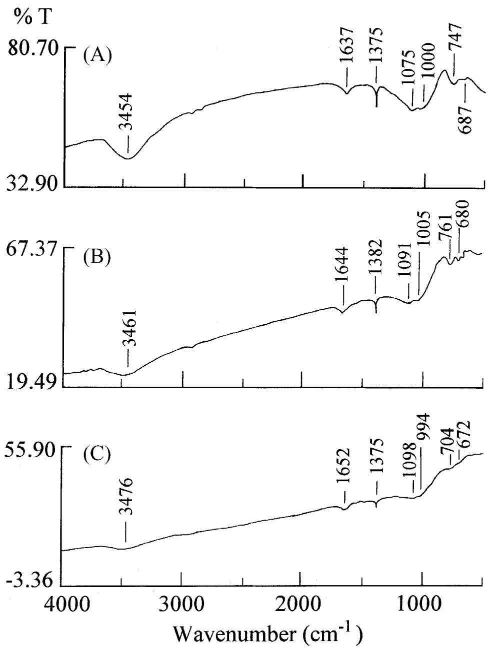

Fig. 2. IR curve of sample heat-treated at 1000 °C (A: sample S1, B: sample S2 and C: sample S3).

that "hydroxy hydrogel" technique may be an ideal process for the development of reactive precursor powder for the ultimate synthesis of any material having engineering and industrial applications. Keeping this in consideration and also considering the engineering importance of low expansion lithium aluminosilicate ceramics, "hydroxy hydrogel" technique in this particular system (Li_2O–Al_2O_3–SiO_2) was implemented in the present studies.

In the present paper, powder precursors in the form of hydroxy hydrogel were prepared for the synthesis of low expansion ceramics to study the relationship between properties and composition of the phases formed in the sintered material.

2. Experimental

2.1. *Preparation of β-spodumene powder precursor*

β-Spodumene powder precursor in the form of hydroxy hydrogel of different compositions in the system Li_2O–Al_2O_3–SiO_2 were prepared by following method as described in an earlier publication [8]. The content of Li_2O, Al_2O_3 and SiO_2 was such that the molar ratio of Li^+:Al^{3+}:Si^{4+} was kept in the ratio of 1:1:2.5, 1:1:3 and 1:1:3.5 so as to develop low expansion β-spodumene ceramics. The powder precursors, dried at 125 ± 5 °C, were calcined at 750 °C for 2 h. The calcined mass was ground and the content of Li_2O, Al_2O_3 and SiO_2 were analyzed chemically. From the analyzed composition, molar ratio of Li_2O:Al_2O_3:SiO_2 was determined and are shown in Table 1.

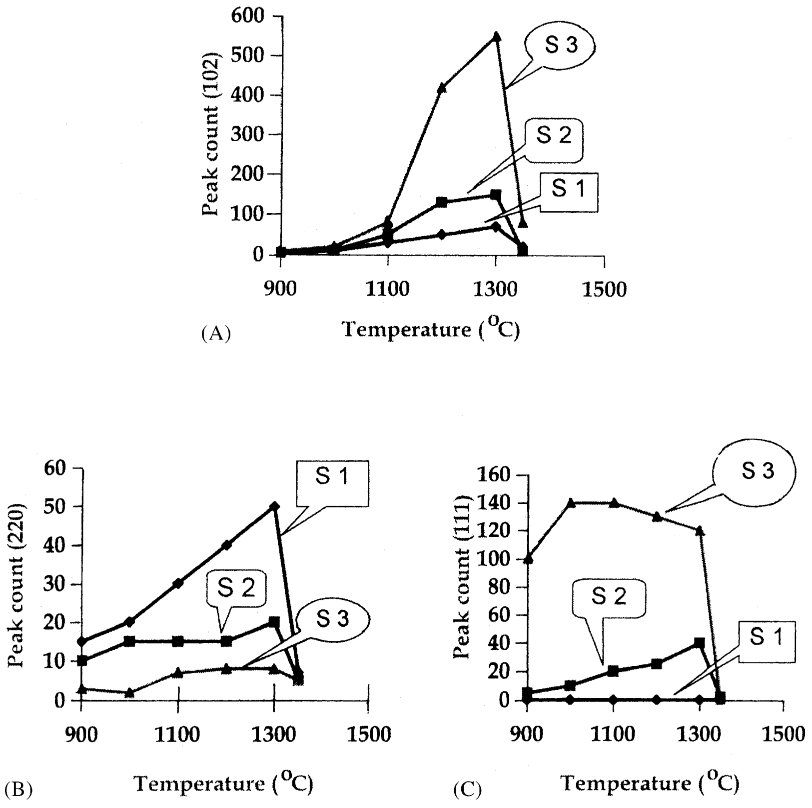

Fig. 3. Formation of different crystalline phases in the sintered compacts at different temperatures of heat treatment. (A) Formation of β-spodumene ($Li_2O{\cdot}Al_2O_3{\cdot}4SiO_2$), (B) formation of lithium aluminate ($LiAl_5O_8$) phase, (C) formation of silica (SiO_2).

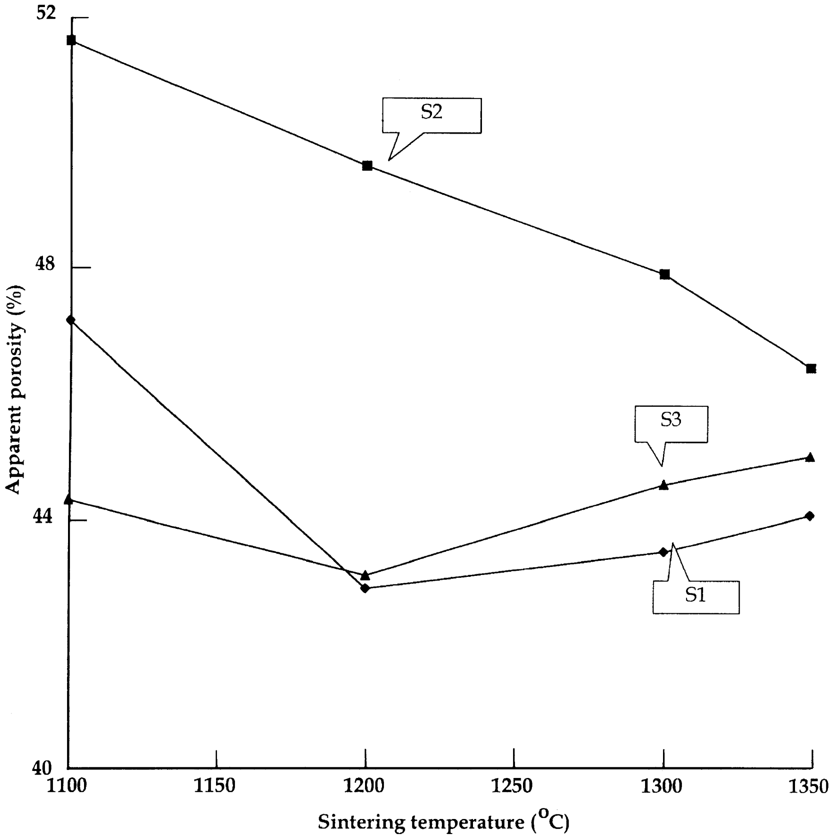

Fig. 4. Apparent porosity (%) of the specimens sintered at different temperatures.

2.2. Preparation of test specimen and sintering

The calcined mass was ground and sieved to pass through 100 mesh B.S. The test specimens were prepared in the form of rectangular bars of dimension 60 mm × 10 mm × 8 mm by uni-axially pressing at 25 MPa followed by cold iso-static pressing at 200 MPa. Sintering experiments were carried out by heating at 1100, 1200, 1300 and 1350 °C in electric furnace in ambient atmosphere with 2 h soaking at the respective peak temperature.

2.3. Measurement of physical properties

The physical properties of the sintered specimens such as fired linear shrinkage, water absorption, apparent porosity and bulk density made from three different compositions were evaluated as per standard procedure [20] and are summarized in Table 2 schematic flow chart showing the preparation of porous low expansion ceramic monolith is illustrated in Fig. 1.

2.4. Measurement of thermal expansion

The sintered specimens for the thermal expansion experiment were cut into bars of dimension 25 mm × 6 mm × 6 mm. The thermal expansion of the specimens was determined in the temperature range 30–800 °C using push rod type Orton Dilatometer (Model 1600D) at heating rate of 10 °C/min in ambient atmosphere.

2.5. IR analysis

The IR study of the specimens heat treated at 1000 °C were done in the range 4000–500 cm^{-1} by means of the Perkin-Elmer model no. 1615 FTIR spectometer using KBr.

2.6. XRD analysis of the powder precursor

The calcined (750 °C) powders which were again fired at 900, 1000, 1100, 1200, 1300 and 1350 °C for 2 h were taken for XRD analysis. The XRD studies for all samples were carried out using a Philips PW-1730 with Cu Kα Radiation and Ni filter.

3. Results and discussion

In an earlier communication [8] it was found that if water is added into a mixture of fumed silica, aluminum nitrate and lithium nitrate, Al^{3+}, Li^{+} and $\text{NO}_3{}^{-}$ ions assume favorable equilibrium position in polymerized silica network structure.

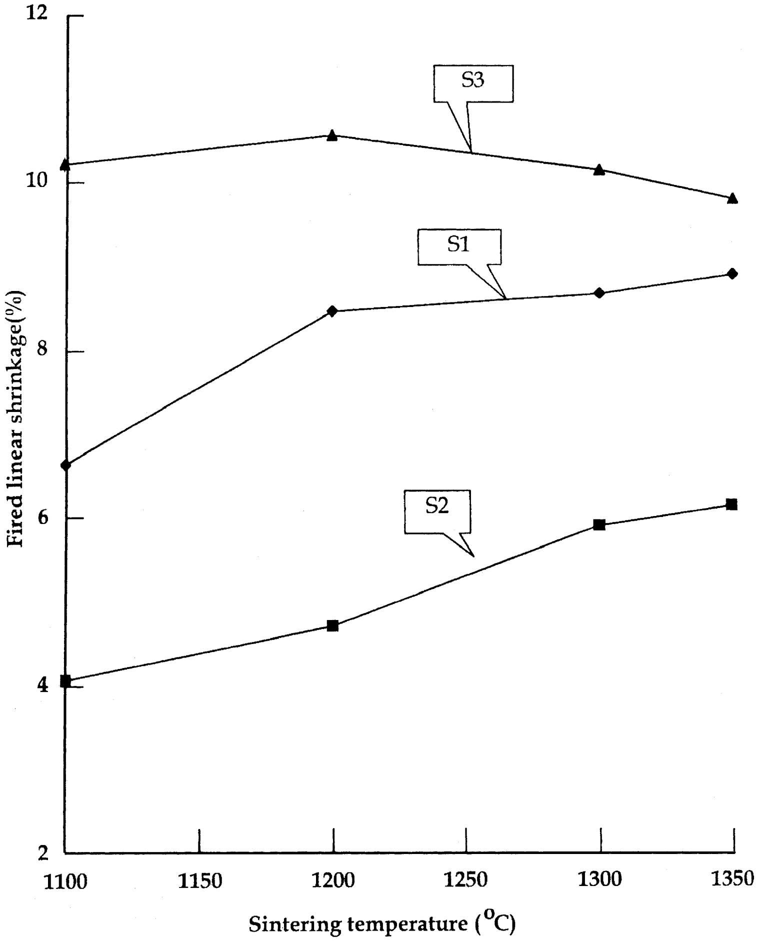

Fig. 5. Fired linear shrinkage (%) of the specimens sintered at different temperatures.

This system during very fast ammonolysis is converted to a gel like mass in which aluminum cation remains partially in octahedral and tetrahedral co-ordination. During dehydration and dehydroxylation processes, water molecules are progressively eliminated leaving behind cavities putting the structure in non-equilibrium state. During gradual heat treatment this structure collapses and a highly reactive powder precursor is formed where traces of hydroxy linkages were still present as was evident from the IR spectral analysis of the samples heat treated at 1000 °C (Fig. 2). The presence of IR absorption bands at 1375 and 1382 cm^{-1} region (Fig. 2) was ascribed to NH^{4+} ion (ammonium nitrate). The retention of this particular peak indicates that all samples contained trace amount of ammonium nitrate even after heat treatment at 1000 °C. The difference in the band intensities between the stretching modes of OH groups and H_2O molecules centered at 3454, 3461 and 3476 cm^{-1} region (presence of hydroxylated water) and the bending mode of H_2O molecules centered at 1637, 1644 and 1652 cm^{-1} region (Fig. 2) is in agreement with the presence of both, H_2O and OH groups. The peaks at frequencies at 3454, 3461 and 3476 cm^{-1} region is contributed mainly by aluminum hydroxyl gel. Therefore, the physical identity of both $Al(OH)_3$ and ammonium nitrate was retained in all samples with varying amounts.

The different phases formed in the specimens after sintering at different temperature were lithium aluminosilicate (β-spodumene) having tetragonal dipyramid crystal structure ($LiAlSi_2O_6$), lithium aluminiumoxide ($LiAl_5O_8$) having cubic crystal structure and silicon oxide (SiO_2) having cubic crystal structure in varying amounts. The peak counts were plotted against the sintering temperature for all samples to determine the relative formation of each phase formed with respect to standard. The formation of different phases at different temperatures was calculated as a function of temperature with the help of XRD analysis. The crystallographic analysis was done with the help of Rietveld method. Profit was used to refine the peak intensity, peak position, peak shape (FWHM) DICVOL and IOT's methods were used to calculate the crystallographic data. Finally Rietveld was used to calculate the relative growth of different phases formed at different sintering temperatures. The relative formation of different crystalline phases at different temperatures of heat treatment are shown in Fig. 3. From the Fig. 3 it is evident that the formation of β-spodumene phase increased with increasing temperature up to 1300 °C

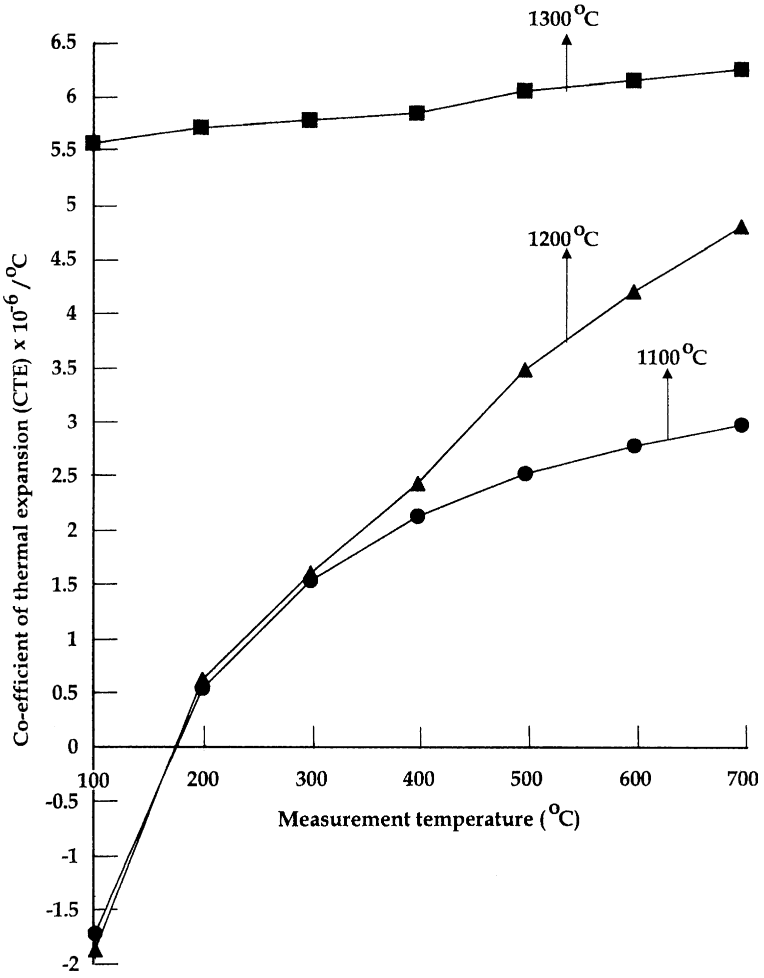

Fig. 6. Thermal expansion behavior in relation to the temperature of measurement of the sample S1 sintered at different temperatures.

which was again followed by a sharp decrease at 1350 °C. At 1300 °C, maximum amount of β-spodumene was formed in sample S3 while much less amount was formed in sample S1. The formation of lithium-aluminate phase increased with increasing heat treatment temperature up to 1300 °C for all samples followed by a sharp decrease at 1350 °C. At 1300 °C, maximum amount of lithium aluminate phase was formed in sample S1 and much less amount was formed in sample S3. The formation of silicon oxide phase increased with increasing heat treatment temperature upto 1300 °C for all the samples followed by a sharp decrease at 1350 °C. At 1300 °C, formation of silicon oxide was more in sample S3 and much lesser amount was noticed in sample S2. There was insignificant formation of silicon oxide phase in sample S1.

Therefore, it may be said that at 1300 °C, lithium aluminate phase was the predominant phase in sample S1 whereas β-spodumene and silicon oxide phases were predominant phases for the sample S3 (Fig. 3). All phases were present in moderate amount in sample S2. The sharp fall of the amount of all the phases in all samples (sintered at 1350 °C) might be due to the initiation of vitrification.

The change of porosities of the samples sintered at four different temperatures (1100, 1200, 1300 and 1350 °C) is shown in Fig. 4. The porosity of the sample S1 and S3 showed slight increase when sintering temperature was raised from 1200 to 1300 °C but in the case of sample S2, a consistent decrease in porosity was indicated. This observation was supported by the behavior of the specimens with respect to dimensional changes occurring at different temperatures as indicated in Fig. 5. This increase in porosity may be due to the elimination of some volatile matter inside the specimens during heat treatment at 1300 °C leaving behind the pores or due to the elimination of low-density phases at this particular temperature, or both.

The thermal expansion behavior of the specimens S1, S2 and S3 sintered at 1100, 1200 and 1300 °C is shown in Figs. 6–8, respectively. It appears from the Figs. 6–8 that coefficient of thermal expansion of the samples S1 and S2, when sintered at 1100 and 1200 °C and measured in the temperature range from room temperature to 100 °C is negative but in case of sample S3, it is positive. The negative thermal expansion is possibly due to the presence of hexag-

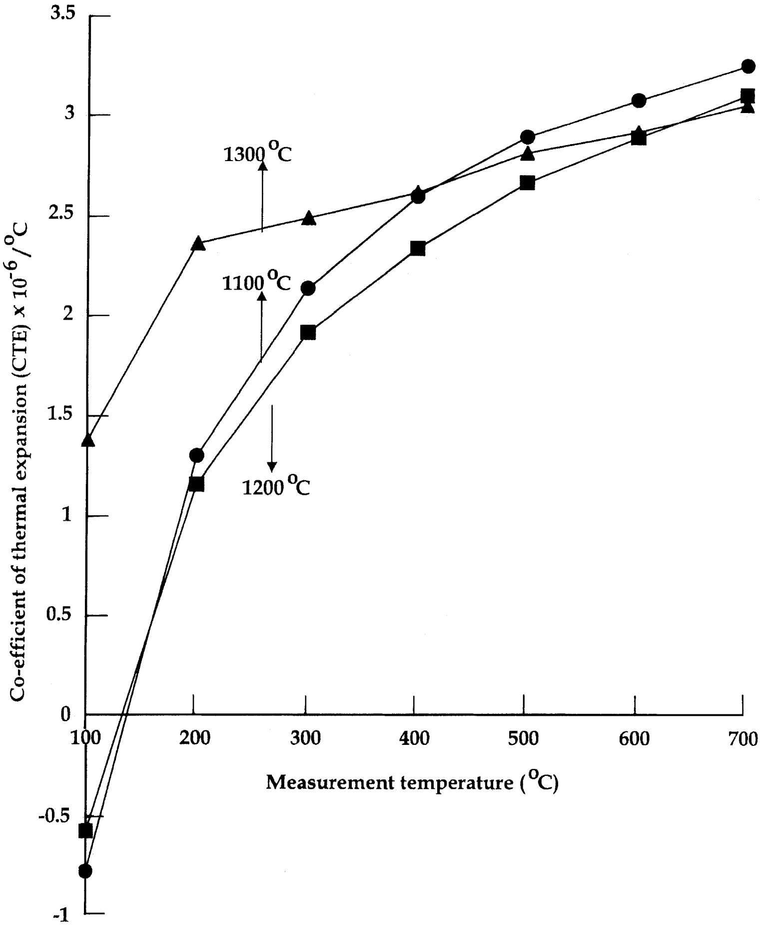

Fig. 7. Thermal expansion behavior in relation to the temperature of measurement of the sample S2 sintered at different temperatures.

onal β-eucryptite (negative thermal expansion contributing phase) as minor phase in samples S2 and S3 when sintered at 1100 and 1200 °C. The positive expansion of sample S3 (sintered at 1100 and 1200 °C) is mainly due to the presence of tetragonal β-spodumene as major phase where no β-eucryptite was detected. In the present study, the sample S1, S2 and S3 have SiO_2 content in increasing order (Table 1). The lower silica content (sample S1) helps the formation of β-eucryptite, lithium aluminum oxide and β-spodumene (Fig. 3), out of which the lithium aluminum oxide phase was found to be dominant. When all samples were sintered at 1300 °C, the initial value of coefficient of thermal expansion measured in the range room temperature to 100 °C is positive but sample S1 showed much higher values of thermal expansion in comparison to S2 and S3 samples. However, at higher temperature of measurement, a consistent increase in thermal expansion values in the range of 100–800 °C was noticed for all samples sintered at 1300 °C.

The sudden rise in the expansion values of the specimen S2 and S3 at around 200 °C (Figs. 7 and 8, respectively) may be possibly due to the presence of cubic crystalline silicon oxide phase as is evident from the Fig. 3C. This phenomenon may be ruled out in the case of sample S1 as the formation of silica phase was insignificant in it.

The sample S1 when sintered at 1300 °C showed the higher thermal expansion values at all temperature in comparison to sample S2 and S3. The low expansion behavior of samples S2 and S3 in comparison to S1 is possibly due to formation of larger amount of β-spodumene (Fig. 3A). Although the formation of silicon oxide phase is low as compared to the formation of β-spodumene in samples S2 and S3 (Fig. 3C), contribution of this phase towards the thermal expansion behavior of the samples S2 and S3 can not be ruled out. Higher thermal expansion behavior of sample S1 (Fig. 6) may be due to the presence of larger amount of lithium aluminium oxide (Fig. 3B) which is the predominant phase in sample S1. Thermal expansion behavior of the samples S2 and S3 were similar and slight higher thermal expansion of sample S3 may be due to the presence of higher silicon oxide (Fig. 3C). Therefore, it may be concluded that the sintered specimen prepared at different temperatures belongs to varying expansion group of materials and this behavior is strongly related to the formation of different phases in the sintered specimens.

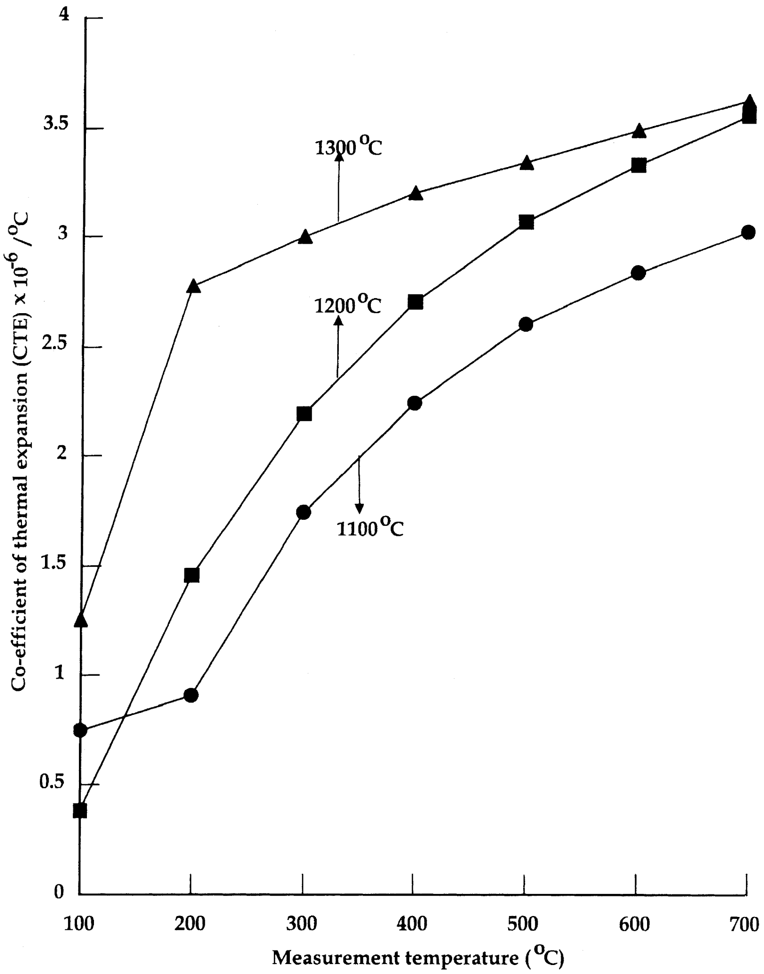

Fig. 8. Thermal expansion behavior in relation to the temperature of measurement of the sample S3 sintered at different temperatures.

4. General discussion

The principle phases in the samples S1, S2 and S3 are lithium aluminium oxide, β-spodumene and silicon oxide in varying amounts. The predominant phase present in the sample S1 is lithium aluminium oxide along with the minor phases like β-eucryptite and β-spodumene. In comparison to S2 and S3 specimens, S1 contains maximum amount of lithium aluminium oxide and lesser amount of β-spodumene (Fig. 3A). Slightly higher density of S1 with respect to samples S2 and S3 (Table 2B) may be due to the presence of maximum amount of lithium aluminium oxide phase (density $= 3.62\,\mathrm{g/cm^3}$) [21–23]. With increasing temperature of heat treatment the density of S1 sample increased from $1.32\,\mathrm{g/cm^3}$ (at 1100 °C) to $1.46\,\mathrm{g/cm^3}$ (at 1350 °C) (Table 2B) indicating the formation of increasing amount of lithium aluminium oxide, which is substantiated by their results of thermal expansion coefficient values, which also increases with increasing temperature of heat treatment of the specimens. For example the coefficient of thermal expansion values the sample S1 sintered at 1100 °C (Fig. 6) and measured at 700 °C is $2.98 \times 10^{-6}\,{}^{\circ}\mathrm{C}^{-1}$ which is increased to $6.27 \times 10^{-6}\,{}^{\circ}\mathrm{C}^{-1}$ when the specimen was heat-treated at 1300 °C (Fig. 6). The similar analysis of the other specimens leads to identical conclusion that both the property, density (Table 2) and coefficient of thermal expansion (Figs. 6–8) are strongly dependent on the phases present in the specimens.

5. Conclusions

Lithium aluminosilicate powder precursor in the hydroxy hydrogel form was found to be suitable for making low expansion ceramics through wet interaction technique.

Sintered specimen prepared at different temperatures belongs to low expansion group of materials and this behavior is strongly related to the different phases in the sintered specimens.

Thermal expansion characteristics and density of the sintered specimen were found to be primarily related to the composition of the phases formed during sintering.

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