Journal of Ovonic Research Vol. 7, No. 3, May - June 2011, p. 61 - 66

EFFECT OF SINTERING TEMPERATURE ON THE DIELECTRIC PROPERTIES OF Li_2SiO_3 CERAMIC

ABHIJIT PRASAD*, AMITABHA BASU, MANOJ KUMAR MAHATA
Department of Applied Physics, Indian School of Mines, Dhanbad-826004, India

Lithium silicate (Li_2SiO_3) ceramic was prepared via solid-state reaction technique. The effects of sintering temperature on the dielectric properties of Lithium Silicate ceramic were investigated. The density was found to be an important factor affecting the dielectric loss of the sample. The optimum sintering temperature of the Lithium Silicate (Li_2SiO_3) ceramic was found to be around 900 ^oC. The dielectric constant (ε_r) and dielectric loss ($\tan\delta$) have been measured at different temperatures for frequencies up to 5 MHz. In the all cases, a strong frequency dispersion of permittivity was observed in the low frequency region followed by a nearly frequency independent behavior above 100 kHz. The sample sintered at 900 ^oC has a high dielectric constant and low dielectric loss in the high frequency region at room temperature. The dielectric loss ($\tan\delta$) of the sample sintered at 900 ^oC was found to be 0.0028 at 3 MHz. From the dielectric studies, we can prescribe Li_2SiO_3 sample as a good dielectric material since it possesses low tangent loss.

(Received May 30, 2011; Accepted June 16, 2011)

Keywords: Ceramics; Dielectrics; Sintering

1. Introduction

Lithium silicate ceramic systems belong to technologically important class of ceramic materials for various applications. Thus, for example, there has been research in recent years on their application as electronic devices, as CO_2 captors and as breeder materials for nuclear fusion reactors, in addition to other more well-known applications such as in batteries and in low thermal expansion glass ceramics used in ceramic hobs [1-10]. Various properties of Lithium silicate ceramics such as dielectric, conductivity and other properties depend on the composition and microstructure. In this study, the investigation is concerned with the preparation of Lithium Silicate (Li_2SiO_3) ceramic and the effect of sintering temperature on the dielectric properties of Lithium silicate ceramic are also investigated. This study also presents the influence of temperature and frequency on the dielectric properties of Lithium silicate ceramic.

2. Experimental

Lithium silicate (Li_2SiO_3) ceramic was prepared via solid-state reaction technique using high purity ingredients: Lithium Carbonate (Li_2CO_3) (99.5%) and Silicon Dioxide (SiO_2) (99.5%) in the following molar ratio:

$$Li_2CO_3:SiO_2 \qquad (1:1)$$

In the first stage, the initial charge was thoroughly mixed in agate mortar for 2 h, including wet mixing in acetone media for 1 h. The mixture was then calcined at 900 ^oC for 3 h. The calcined powder was then cold pressed into cylindrical pellets of 12 mm diameter and 2 - 3 mm of thickness at a pressure of 80 MPa using a hydraulic press. PVA (poly vinyl alcohol) was used as a

---

[*] Corresponding author: abhijit_133@rediffmail.com

binder for preparing pellets. These pellets were then sintered at 800 °C, 850 °C, 900 °C and 950 °C for 2 h in an air atmosphere.

The compound formation was confirmed by X-ray diffraction (XRD) using Philips Xpert-pro X-ray powder diffractometer in a wide range of the Bragg angles 2θ ($10^o \leq 2\theta \leq 90^o$) being irradiated by Co (λ = 1.78897 Å). The surface morphology was recorded using Field emission scanning electron microscope JEOL (model: JSM – 5800F). In order to study the dielectric properties of the compound, both the flat surfaces of the samples were polished and electroded with air-drying conducting silver paint. After electroding, the pellets were dried at 150 °C for 2 h to remove moisture, if any, and then cooled to room temperature before taking any electrical measurement. The dielectric permittivity (ε_r) and loss tangent (tanδ) of the sample was measured using an impedance analyzer (HIOKI 3532 LCR Hi-TESTER) in the frequency range (100 Hz – 5 MHz) at some selected temperatures (35 °C, 50 °C, 100 °C, 150 °C, 200 °C, 250 °C, 300 °C & 350 °C).

3. Results and discussion

3.1. Structural

Fig.1 shows the bulk densities of the Li_2SiO_3 ceramic sintered at various temperatures for 2 h. The density of the Li_2SiO_3 ceramic was initially increased with sintering temperature up to 900 °C but decreases at 950 °C. We can draw the conclusion that Li_2SiO_3 ceramic could be well-sintered at 900 °C.

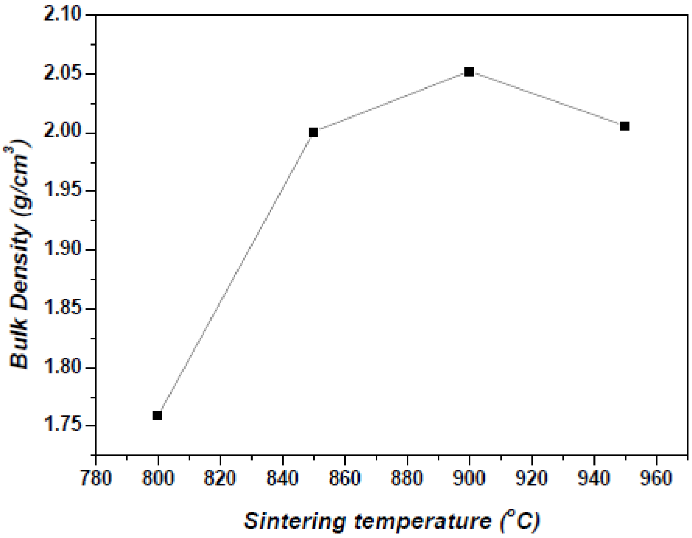

Fig. 1. Variation of bulk density of Li_2SiO_3 ceramic with sintering temperature.

XRD pattern of Lithium silicate (Li_2SiO_3) ceramic sintered at 900 °C is shown in Fig. 2. Detailed structural analysis in different crystal structure and cell constants exhibit that the sample has an orthorhombic structure with lattice parameters: a = 9.396 Å, b = 5.396 Å, c = 4.661 Å, which is in well agreement with the phase given in JCPDS card number 83 – 1517. The crystallite size of the powder sample was roughly estimated from broadening of reflection peaks using Scherrer's equation [11], $D = 0.89\lambda / (\beta_{1/2}\cos\theta)$, where λ = 1.78897 Å and $\beta_{1/2}$ = half peak width of reflections. The average crystallite size was found to be ~ 30 nm. Other effects of the broadening were ignored.

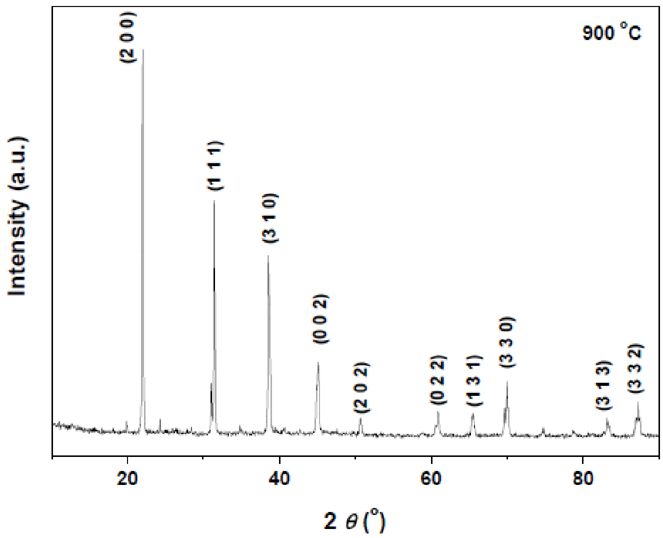

Fig. 2. Room temperature XRD pattern of Lithium silicate (Li_2SiO_3) ceramic sintered at 900 °C.

The FE-SEM micrograph was taken on the fractured surface of the sample using scanning electron microscope. The highly distinctive, more or less uniform and compact grain distributions (with less voids) are observed. It shows the polycrystalline texture of the material. The average grain size of the sample sintered at 900 °C was found to be 2-4 μm. There are a few islands and holes in SEM, which suggests that the pellet sample was of high density. Fig. 3 shows dense and homogeneous microstructure of Lithium silicate (Li_2SiO_3) ceramic sintered at 900 °C.

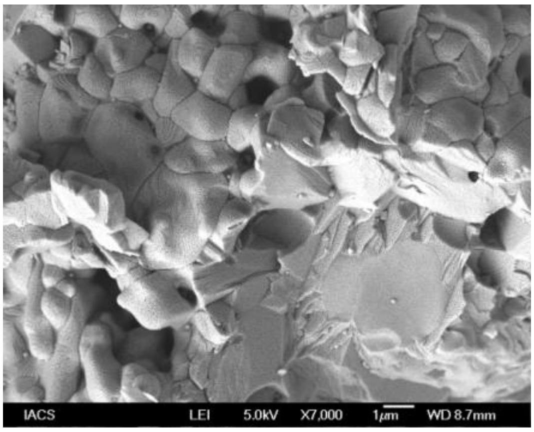

Fig. 3. FE-SEM of lithium silicate (Li_2SiO_3) ceramic sintered at 900 °C.

3.2. Dielectric properties

Fig. 4 show the variation of dielectric constant (ε_r) and loss tangent ($\tan\delta$) with frequency for Li_2SiO_3 ceramic sintered at three different temperatures. In all cases both the parameters decrease on increasing frequency indicating a normal behavior of dielectric materials having mobile charge carriers (i.e., ions and electrons). The fall in dielectric constant arises from the fact that the polarization does not occur instantaneously with the application of the applied electric field as charges possess inertia. The delay in response towards the impressed alternating electric field leads to loss and hence decline in dielectric constant. The loss tangent ($\tan\delta$) also decreases with increasing frequency. It is observed that at higher frequency, these parameters became almost frequency independent. It is also observed that at high temperatures (> 250 °C), width of the plateau region decreases with the increase of sintering temperature. The sample sintered at 900 °C has a low dielectric loss at high temperatures. The frequency dependence of loss tangent exhibits interesting results. It is evident from Fig. 4 that at high temperatures (> 300 ^oC), the loss tangent reaches the instrumental saturation value (tanδ = 10) in the low frequency region but at high frequency (i.e. 1 MHz) the value of tanδ drops down from this saturation drastically. It shows the possibility of using the material for high frequency applications with low dissipation factor.

Fig.4. Variation of dielectric constant (ε_r) and loss tangent (tanδ) with frequency for the samples sintered at (a)800 ^oC (b)850 ^oC and (c)900 ^oC.

Table 1 shows the room temperature ε_r and tanδ values of the samples measured at 1 MHz. The dielectric constant of the samples is in the range of 15.13 – 18.51.

Table 1: Physical properties of Li_2SiO_3 ceramic sintered at different temperatures.

(a) **(b)**

Fig.5 (a) variation of dielectric constant (ε_r) of all the samples with frequency at room temperature (35^oC) (b) variation of loss tangent (tanδ) with sintering temperature at 3MHz.

The sample sintered at 900 ^oC for 2 h have the lowest dielectric loss at 3 MHz (Fig. 5 (b)) and high dielectric constant (Fig. 5 (a)) at room temperature. It is clear that the sample sintered at 900 ^oC has the best dielectric property.

4. Conclusions

The Li_2SiO_3 ceramic was prepared by a solid-state reaction technique. X-ray structural study reveals an orthorhombic crystal structure of the material. The optimum sintering temperature for Li_2SiO_3 ceramic was found to be 900 ^oC. The surface morphology of the compound is studied through FE-SEM, which shows the uniform distribution of well compacted grains throughout the sample. Dielectric constant and dissipation factor decrease with the increase in frequency. The sample sintered at 900 ^oC has the best dielectric property.

References

[1] C. A. Vincent, Solid State Ionics, **134**, (1-2), 159 (2000).
[2] C.-H. Lu and L.Wei-Cheng, Journal of Materials Chemistry, **10**(6), 1403 (2000).
[3] M. Broussely, F. Perton, P. Biensan, et al., Journal of Power Sources, **54**, (1), 109 (1995).
[4] V. Subramanian, C. L. Chen, H. S. Chou, G. T. K. Fey, Journal of Materials Chemistry, **11**(12), 3348 (2001).
[5] X. Yang,W. Tang, H. Kanoh, K. Ooi, Journal ofMaterials Chemistry, **9**(10), 2683 (1999).
[6] H. Kudo, K. Okuno, and S. O'Hira, Journal of Nuclear Materials, **155–157(**1), 524 (1988).
[7] H. Pfeiffer, P. Bosch, and S. Bulbulian, Journal of Materials Chemistry **10**(5), 1255 (2000).
[8] H. Pfeiffer and P. Bosch, Chemistry of Materials, **17**(7), 1704 (2005).
[9] H. A. Mosqueda, C. Vazquez, P. Bosch, and H. Pfeiffer, Chemistry of Materials, vol. 18, no. 9, pp. 2307–2310, 2006.
[10] H. Pfeiffer, C. V´azquez, V. H. Lara, and P. Bosch, Chemistry of Materials, **19**(4), 922 (2007).
[11] P. Scherrer, Gö ttinger Nachrichten **2**, 98 (1918).