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Characterization of Phase formation and Microstructural Evolution on Dielectric Properties of $SrO-B_2O_3-SiO_2$ and $BaO-SrO-Nb_2O_5-B_2O_3-SiO_2$ glasses

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ABSTRACT. The phase formation, microstructural evolution and dielectric properties of $SrO-B_2O_3-SiO_2$ (SBS) and $BaO-SrO-Nb_2O_5-B_2O_3-SiO_2$ (BSNBS) glasses were studied. SBS and BSNBS glasses were prepared via solid state reaction method. The raw materials of SBS and BSNBS were wet mixed for 24 hours and then dried overnight in oven. The mixing powder of SBS and BSNBS were heated until 1250 °C and 1450 °C, respectively for 2 hours for making a glass. The glass frits was crushed to form a fine powder. SBS and BSNBS glass powder was compacted at 250 MPa and then were heat treated at 800 °C and 650 °C, for 3 hours respectively. X-ray diffractometer (XRD) analysis showed the amorphous phase for both glasses powder after melting process, whereas the formation of $Ba_{0.39}Sr_{0.61}Nb_2O_6$, SiO_2 and Nb_2O_5 phases, respectively were obtained by BSNBS sample after heat treatment process. Observation on scanning electron microscopy (SEM) micrographs showed no grain was observed for SBS sample while fine grain was seen for BSNBS sample. Both dielectric constant and dielectric loss of BSNBS glass was higher compared to the SBS glass.

Keywords: *Glass, Nucleating agent, Phase formation, Microstructure, Dielectric properties;*

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1. INTRODUCTION

The dielectric materials with high energy storage density which can effectively minimize the volume and size of the capacitor are required for high-voltage capacitor application [1]. In order to achieve that, characteristics such as high dielectric constant, low dielectric loss and high breakdown strength for materials are needed [2]. Previously, there were many ceramic materials that have been used as capacitor such as $BaTiO_3$, $MgTiO_3$ and $CaCu_3Ti_4O_{12}$ ceramics [3,4]. However, the pores created during sintering process bring major causes of structural defects, thus result in the degradation of their properties and failure of the devices. After that, glass ceramics materials were introduced where these materials have their advantages which free of porosity, and thus, their electrical breakdown strength might be higher than conventional ceramic dielectric materials [5-7]. They also may have high dielectric constant which is from the precipitated ceramic phases.Dielectric properties (dielectric constant and dielectric loss) of dielectric materials were always

related to the density of bulk body [8]. Even though dielectric constant improves linearly with the increase of density, suitable crystallization phase is more effective to improve the dielectric constant of glass. Therefore, improving the crystallization properties of glass has been attracted much interest. Hence, it is necessary to introduce some nucleating agents for promoting crystallization to the glass. Efficient nucleation of crystals would result in fine-grained microstructures and consequently high storage.

Even though there were researches on fabrication of glass ceramics as high energy storage density capacitor [5-7], there were still lack of studies available in the literature focusing on the correlation between crystalline/amorphous phase formation with the microstructure and dielectric properties of glass. The aim of this study is primary investigation on effect of nucleating agents (Nb_2O_5) to the crystallization phase formation, microstructures, and dielectric properties of SrO-B_2O_3-SiO_2 (SBS) glass in order to obtain the material with high storage density.

2. MATERIALS AND METHODS

SBS glass and BSNSB glass ceramics were prepared via solid state reaction method. The raw materials of BaO (Sigma Aldrich, >99%), SrO (Sigma Aldrich, >99%), Nb_2O_5 (MERCK, >99%), B_2O_3 (Sigma Aldrich, >99%) and SiO_2 (Sibelco, >99%) powder were used for mixing process where SBS glass with compositions of $30.8SrO$-$58.9B_2O_3$-$10.8SiO_2$ (mol%) was wet mixed for 24 hours while BSNBS glass with compositions of $21BaO$-$21SrO$-$18Nb_2O_5$-$10B_2O_3$-$30SiO_2$ (mol%) was wet mixed using the same procedures. Each mixture was dried overnight in oven. BSNBS and SBS mixtures then were melted in alumina crucible using glass melting furnace (Lenton EHF 1800) at 1450 °C and 1250 °C for 2 hours, respectively.

The molten glasses were manually poured into distilled water and the glass frits then were ball milled to form a fine glass powders. Each glass powder was pressed into pellets under a pressure of 250 MPa. The SBS and BSNBS glasses pellets then were heat treated in air at temperature at 800 °C and 650 °C, for 3 hours, respectively. The phase formation of the SBS and BSNBS glasses powder and pellets were analyzed using X-ray diffraction (XRD) (model: Bruker Advance D8) while Scanning electron microscopy (SEM) (model: TM 3000) was used to observe the microstructure of the pellets samples. For electrical measurement analysis, the dielectric properties of the samples were measured using impedance analyzer machine (model: 4291B Hewlett Packard) over a frequency range from 1 MHz to 1 GHz.

3. RESULTS AND DISCUSSIONS

Fig. 1 shows the XRD pattern of SBS and BSNBS glasses powder after melting glass process. As shown in the figure, there were no peaks shown which indicates the complete amorphous phase for each glass after melting process. Meanwhile, Fig. 2 shows the XRD diffragtograms of the as- devitrified SBS and BSNBS glasses samples after heat treatment process. XRD results shows a complete amorphous phase also can be seen for SBS glass whereas the crystalline phases formed for BSNBS glass ceramics.

For BSNBS glass ceramics sample, a main $Ba_{0.39}Sr_{0.61}Nb_2O_6$ phase (ICDD Data File Card No. 01-088-0785) with secondary phases of SiO_2 (ICDD Data File Card No. 00-043-0784) and Nb_2O_5 (ICDD Data File Card No. 00-034-0898) phases were identified. The formation of crystalline phases for BSNBS glass sample is due to the addition of Nb_2O_5 where this material is known as nucleating agent for glass making process [9].

The dielectric properties of SBS and BSNBS glasses samples were plotted in the Fig.4. Based on dielectric constant results shown in Fig. 4(a), the dielectric constant of BSNBS glass sample were higher over the frequency range from 1 MHz to 1 GHz compared to the dielectric constant of SBS glass sample. At 1 MHz, the dielectric constant of BSNBS glass sample was 18 and the value was consistent until 1 GHz. Meanwhile, SBS glass sample also maintained the dielectric constant of 6 over that frequency range. It shows that the grain and grain boundaries of BSNBS glass sample play a big part for contributing the high dielectric constant compared to SBS glass sample

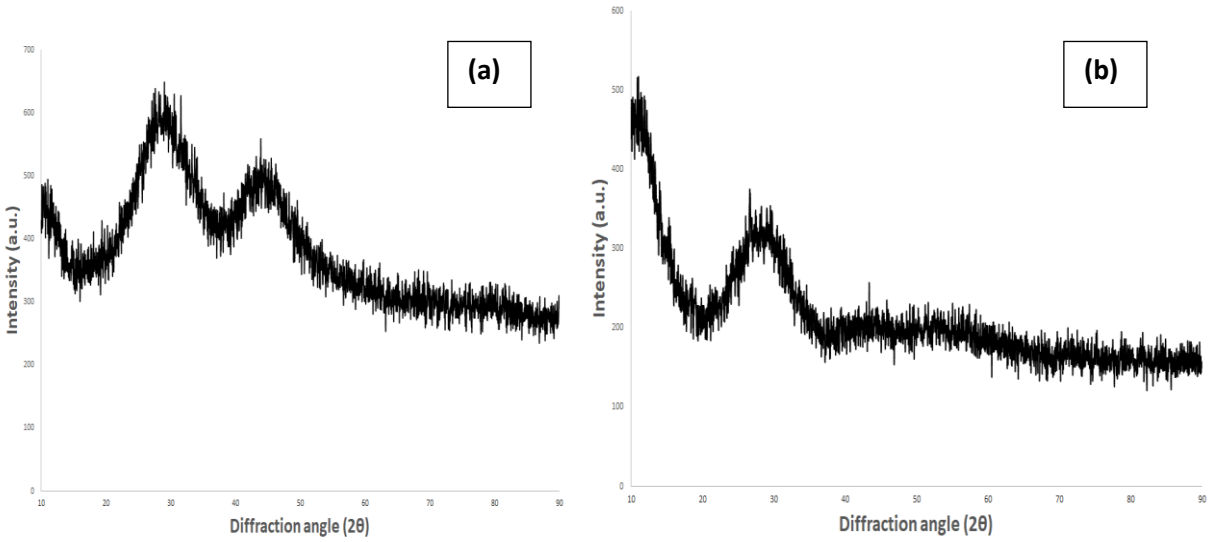

Fig. 1 XRD pattern of (a) SBS and (b) BSNBS glasses powder

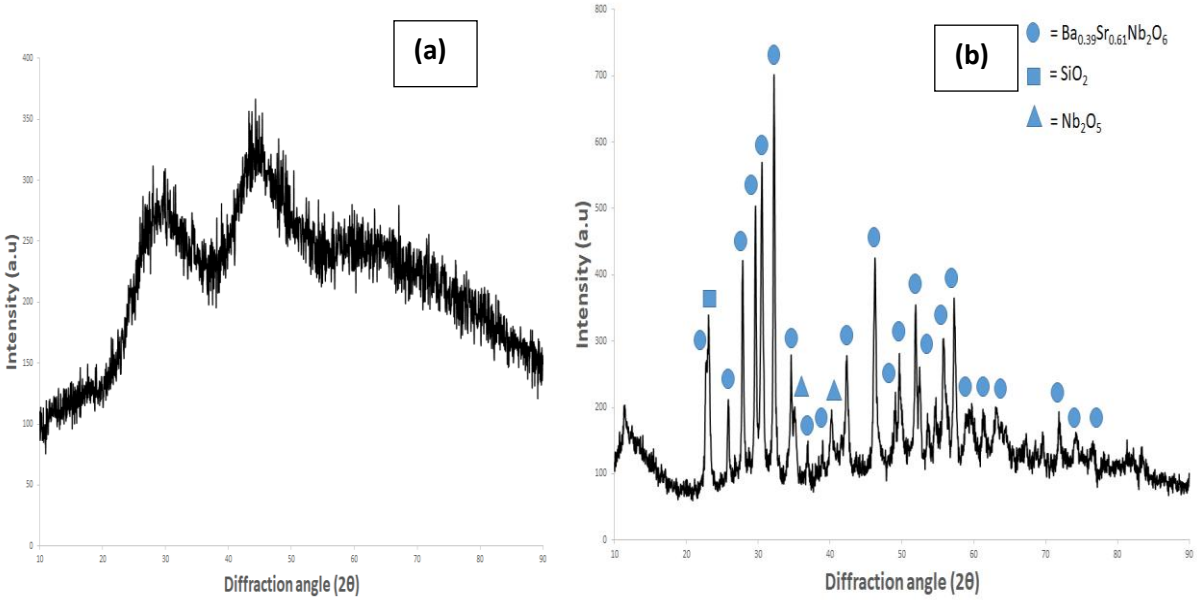

Fig. 2 XRD pattern of heat treated of (a) SBS and (b) BSNBS glasses sample

Fig. 3 shows the microstructure of the SBS and BSNBS glasses after heat treatment process. It can be observed that the microstructure of these glasses were quite different. In Fig. 3(a), smooth surface can be seen clearly for SBS glass sample. Unlike SBS glass which was not crystallized during heat treatment process, crystallized grain of ~0.5 μm can be observed on the surface of BSNBS glass sample (Fig. 3(b)). The addition of Nb_2O_5 as nucleating agent contributed to the presence of crystallization phase, thus promote the grain growth of the glass [9].

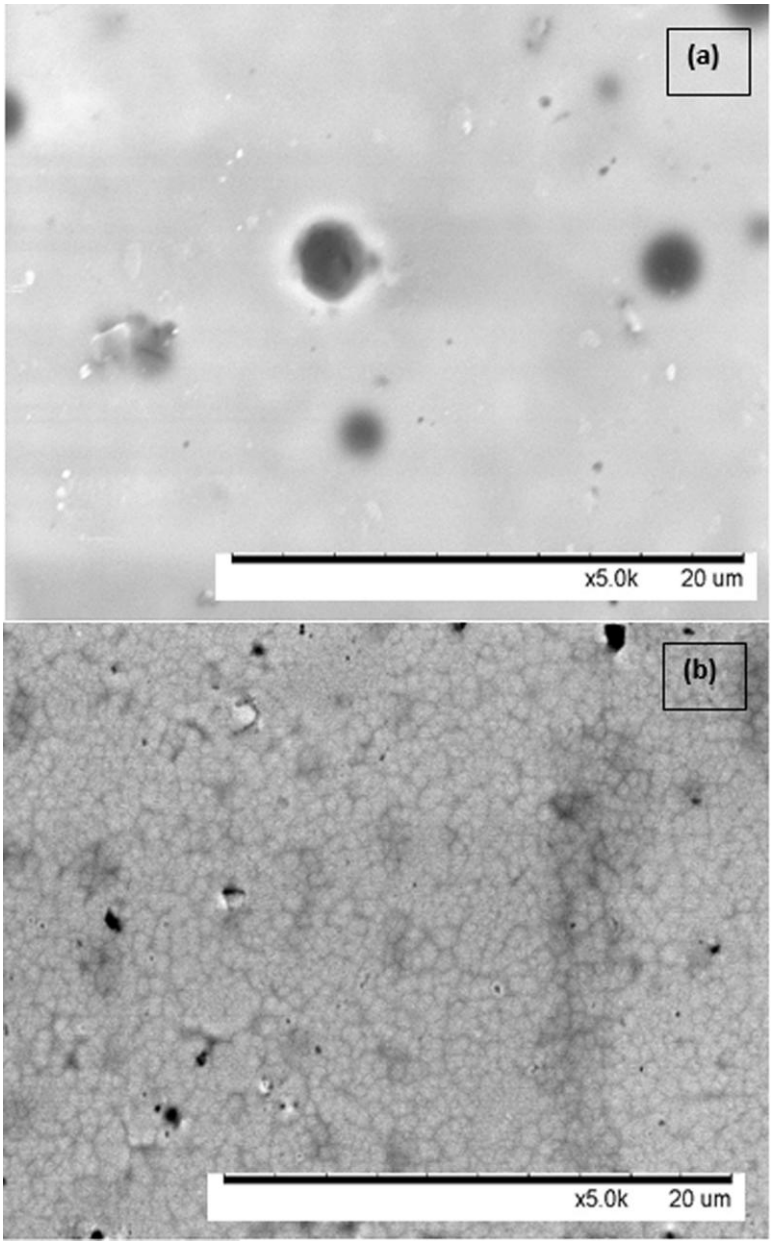

Fig. 3 SEM micrographs of as devitrified of (a) SBS and (b) BSNBS glasses

According to Ab Rahman et al. [10], they stated that high dielectric constant was attributed to high density bulk body and also the average grain size of materials. Thus, the grain that growth for BSNBS glass ceramics after heat treatment process contributed to the high dielectric constant of this material.

Besides, the dielectric loss of BSNBS glass also higher than dielectric loss of SBS glass. At 1 MHz, the dielectric loss of SBS and BSNSB glasses were 0.009 and 0.007, respectively. While the dielectric loss of SBS and BSNBS glasses were 0.02 and 0.001 at 1 GHz, respectively. Higher dielectric constant of BSNBS contributed to the higher dielectric loss of this glass [11,12]. Thus, it is apparent that there were exists a good connectivity between the crystallite/amorphous phase and crystallite grain growth to the dielectric properties of the glasses.

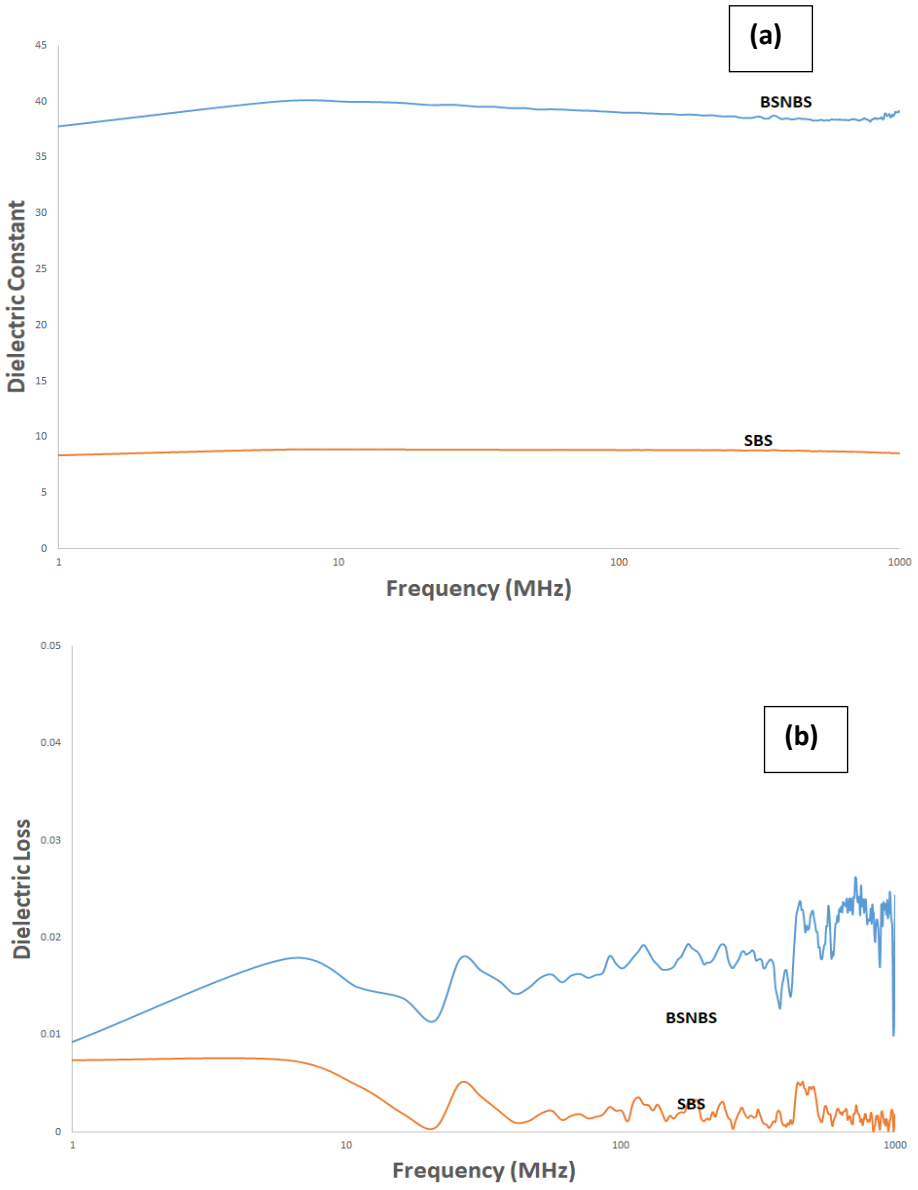

Fig. 4 Dielectric properties of SBS and BSNBS glasses (a) dielectric constant and (b) dielectric loss

4. SUMMARY

SBS and BSNBS glasses were successfully prepared via solid state reaction method. From the XRD analysis, amorphous phase was observed for SBS sample while the formation of $Ba_{0.39}Sr_{0.61}Nb_2O_6$ phase alongside with SiO_2 and Nb_2O_5 phases were seen for BSNBS sample after heat treatment process. SEM micrographs show that smooth surface were seen for SBS glass while fine grain were observed for BSNBS glass. The dielectric constant and dielectric loss of BSNBS glass were higher compared to dielectric constant and dielectric loss of SBS glass. Thus, it shows that the nucleating agent (Nb_2O_5) addition can change the phase formation from amorphous to crystalline phase, induce the grain growth and increase the dielectric constant of glass.

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