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Study of Glasses in Boro Aluminosilicate System.

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ABSTRACT

Two glass compositions 25 Ba_2CO_3-25 Sr_2CO_3-25 B_2O_3-20 SiO_2-5 Al_2O_3 (SBBSA25) and 50 Sr_2CO_3-25 B_2O_3-20 SiO_2-5 Al_2O_3 (SBBSA0) are prepared by normal melt quenching technique. The glasses are characterized by X-ray Diffraction technique (XRD) and Fourier Transform Infrared Spectroscopy(FTIR). Density measurements of the glasses are done using Archimedes method. Heat treatment by controlled crystallization method at 875^{o}C revealed evolution of crystalline phases in SBBSA0 system. The glass systems prepared can be further investigated with regard to their use in SOFC applications as sealants.

Key words: SOFC, glass, glass ceramic, sealants, XRD, FTIR.

I. INTRODUCTION

Depletion of fuel deposits is a developing threat for the present and future generations. Moreover the concern for humans towards protecting nature from harmful outcomes of burning fuels has also being increased nowadays and so research works are progressing in full swing concentrating in the development of least hazardous fuel cells . Fuel cells are getting attraction because of their environmental friendliness. Among the fuel cells, Solid Oxide Fuel Cells(SOFC´s) hold practical promise over other types of fuel cells because of their all-solid construction, and most importantly, that they offer the promise of far more efficient conversion of our dwindling fossil fuel supplies into electricity with up to 65% efficiency for electric power and 85% efficient for cogeneration[1-4].

Solid Oxide Fuel Cells (SOFC) converts the chemical energy of fuels such as hydrogen into electricity by electro chemical oxidation of fuel at 700-1000^{o}C. In general the SOFCs are mainly two types, Planar SOFC and Tubular SOFC depending on the geometry. Of these, planar design is the most promising due to easier fabrication, improved performance and relatively high power density. In planar SOFC which involves stacking of many unit cells(anode/electrolyte/cathode) separated by metallic interconnect plates, seal is an important criteria. Good seals prevent fuel leakage and air mixing at high temperature. Typical conditions under which the seal will be exposed include high operating temperature (700-800^0 C), varying partial pressure (air and fuel), problems of chemical compatibility with other fuel cell components etc. Seals also has to be robust enough to withstand numerous thermal cycles and reliable enough to sustain long term operation at high temperature. The most common sealants for SOFC are glass ceramic materials and these have been shown to operate in fuel cells for more than 10000 h with no significant degradation. But there are not many universally accepted/applicable seal concepts.[1-7]. The development of sealants for SOFC present a significant challenge as they have to meet these very restrictive requirements as detailed above.

Glasses and glass ceramics has been recognized to bond well with metals and alloys and so they have been developed and studied for their application as sealants in SOFC´s. Many glasses and glass ceramics with attractive properties for use as sealants are already known in literature, but with properties varying in different working conditions. The development of novel glass ceramic with advanced properties is therefore considered among main challenges in this field.

In the light of the above mentioned perspectives the main objective of this work is designing novel glass and glass ceramics with apt functional properties which is applicable for intermediate temperature SOFC operating atmosphere. Aluminosilicate glasses are thoroughly being investigated by researchers worldwide for their use as sealants in SOFC applications. In the present work we have chosen Strontium-Barium-Boro Alumino Silicate system for study. In this study we developed two batch glasses with varying strontium to barium ratio and discussed the structural and physical properties.

2. Materials and Methods

Each batch of the glass were designed by taking stoichiometric amounts of different constituent oxides and carbonates of 99.9% purity. The desired weight fraction of the ingredients were mixed in an agate mortar using pestle with distilled water as medium. The conventional melt quench technique was followed in glass preparation. The compositions were melted in an alumina crucible at 1280^oC for 2 hours in air. The molten glass were quenched on a preheated iron plate. The obtained glasses were immediately annealed at 350^oC for 2 hour to prevent thermal stresses.

The chemical composition of the glasses are listed in Table1.

Name of samples	Ba_2CO_3	Sr_2CO_3	B_2O_3	SiO_2	Al_2O_3
SBBSA25	25wt%	25wt%	25wt%	20wt%	5wt%
SBBSA0	nil	50wt%	25wt%	20wt%	5wt%

Reagent grade Ba_2CO_3, Sr_2CO_3, B_2O_3, SiO_2, Al_2O_3 from Sigma Aldrich were used as starting materials. The composition of the boroaluminosilicate is maintained as 25wt%B_2O_3, 20wt%$Si0_2$, 5wt%Al_20_3.

2.2 Characterization Techniques

The evolution of phases in glass powder samples were monitored by Powder X ray Diffraction technique. The XRD data were collected at room temperature. XRD data of heat treated glass samples were also taken. Archimedes method was employed to measure apparent density of the bulk glass. Fourier Transform Infrared Spectroscopic studies were carried out in glass samples to identify the functional groups present in the sample. The infrared spectra help us to identify the compounds present and type of stretching.

3. Results and Discussions

3.1 Glass Forming Ability

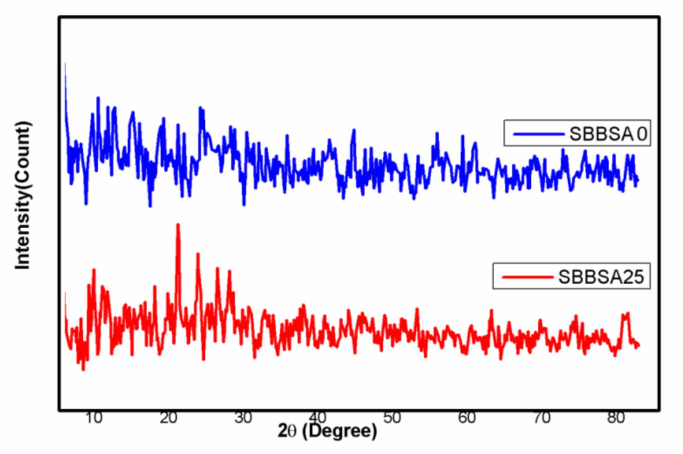

Figure 1. XRD patterns of as prepared glass samples

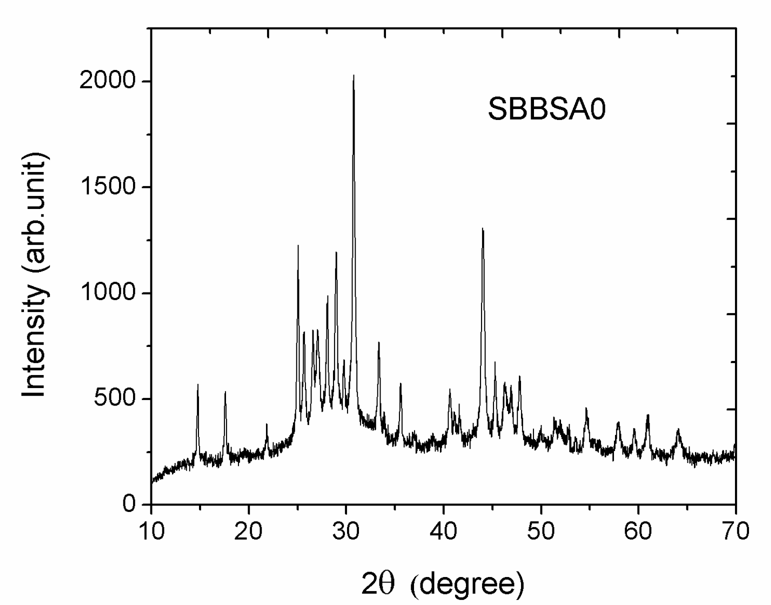

Figure 2. XRD pattern of the heat treated sample SBBSA0 at 875^{o} C

The XRD patterns of the melt quenched glasses are shown in Fig1. The amorphous nature of the glasses are confirmed from XRD analysis. The glass composition named SBBSA0 after heat treatment at 875^{o}C was powdered and the evolution of crystalline phases was confirmed in XRD analysis (Fig2),whereas glass composition SBBSA25 after heat treatment at 750^{o}Cdidnt showed up with XRD pattern resembling crystalline phase.

4.2 Glass Properties

The relative density of the prepared glasses are calculated using the data obtained from Archimedes method and is calculated using the equation given below.

$$RD = \frac{W_{\text{air}}}{W_{\text{air}} - W_{\text{water}}}$$

The density measurements yielded 3.62 g/cc for SBBSA0 and 3.85 g/cc for SBBSA25 glasses respectively.

4.3 Structure of glass

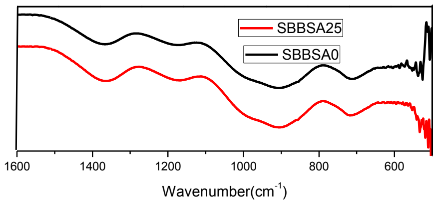

Fig 3 FTIR spectra of SBBSA25 and SBBSA0 glasses.

The room temperature FTIR transmittance spectra of the SBBSA25 and SBBSA0 glasses are shown in Figure3. The glass exhibits three broad transmittance bands in the region 600-1600cm^{-1}. The lack of sharp feature is an indicative of general disorder in the silicate network mainly due to the wide distribution of Q^n units (polymerization in the glass stucture, where n denotes the number of bridging oxygens) occurring in the glasses. The most intense band in the 800-1200cm^{-1} region indicates the Si-O-Si linkages in the SiO_4 tetrahedron unit. The appearance of high intensity broad band at 953cm^{-1} due to the non bridging Si-O terminal stretching vibrations, suggests that Q^2 units are highly localized. With respect to the aluminium co ordination in glass structure, the presence of transmittance band from medium to strong intensity in the 600-700cm^{-1} region is atypical feature of stretching vibration of Al-O bond with aluminium ions in all four coordination.

Table 3 shows band assignments typical to band peaks/wave numbers.

Band peaks, Wave number (cm^{-1})	**Band Assignments**
1355 - 1370	Vibrations of metal cation Ba^{2+}.
930 - 940	Bonding of B-O-B linkages (diborate linkage)

678 - 692	the diborate linkage B-O-B, in the borate glassy network
564	stretching vibration of B-O-Si linkage.
512	non-bridging Si–O stretching vibrations.

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