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Properties of silver phosphate glass doped with nanosize zinc oxide

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ABSTRACT

High valence cations when doped in phosphate glasses, modify the properties, basically due to changes in glass structural network, through formation of cross-linked bonds. In the present paper 5, 10, 15 and 20 wt% nanosize ZnO were doped in Ag_2O–P_2O_5 glass prepared by melt-quench method. Glass transition temperatures were determined by DSC technique, whereas structural investigations were carried out by FTIR spectroscopic technique. X-ray diffraction studies showed that all the prepared glasses are poorly crystalline. SEM studies were made to have an idea about morphological changes in the presence of nanosize ZnO. Electrical conductivities and dielectric constants were measured as a function of temperature and frequency. Cole–Cole plot indicated that grain boundary and relaxation processes affected the electrical properties. It was found that glass transition temperatures increased in the presence of ZnO and conductivities increased with frequency.

1. Introduction

When designing new glass compositions, nearly the whole periodic table is available to serve as the palette of possible ingredients. Phosphate glasses have number of properties which make them technologically important [1–4]. Phosphate glasses are important materials because of their structural versatility to accept a large number of cations and/or anions [5]. The structure of these glasses can be described as a network of PO_4 structural units linked through covalent bonding of corner shared oxygen atoms, referred to as bridging oxygen atoms (BOs). The addition of network modifying oxides in the glass, leads to the depolymerization of the phosphate chains by P–BO–P bonds, and subsequent formation of nonbridging oxygen ions (NBOs). Therefore, the linked phosphate tetrahedral can have three(Q^3), two(Q^2), one(Q^1) or none bridging oxygen ions (Q^0)[5]. Further the addition of alkaline earth metal ions increase the chemical stabilities of phosphate glasses [6,7]. Recently we studied the effect of SrO doping in silver phosphate glass on structural and electrical properties and found interesting results [8].

In this paper we present a systematic study on dielectric properties, e.g. dielectric constant, dielectric loss and electrical conductivity (over a wide range of frequency and temperature) of nanosize ZnO doped silver phosphate glasses. DSC studies have been made to find out the T_g values. Structural elucidation was done by FT-IR spectroscopic studies.

2. Experimental

2.1. Materials

Silver Nitrate ($AgNO_3$), Ammonium dihydrogen phosphate ($NH_4H_2PO_4$), Zinc Sulfate ($ZnSO_4$) and Sodium hydroxide (NaOH) were used for the preparation of glasses. All the chemicals were of more than 98% purity.

2.2. Methods

2.2.1. Preparation of nanosize zinc oxide

Zinc sulfate was dissolved in distilled water. A dilute solution of sodium hydroxide was added drop by drop till complete precipitation of zinc hydroxide occurred. This was continuously stirred for 4 h in order to avoid agglomeration. After the precipitation, the solution was kept to settle for overnight and the supernatant solution was filtered carefully. The obtained zinc hydroxide was washed with hot water several times and dried in hot air oven at 80 °C. Dried sample was heated at 500 °C for 3 h where nanosize zinc oxide was formed [9,10].

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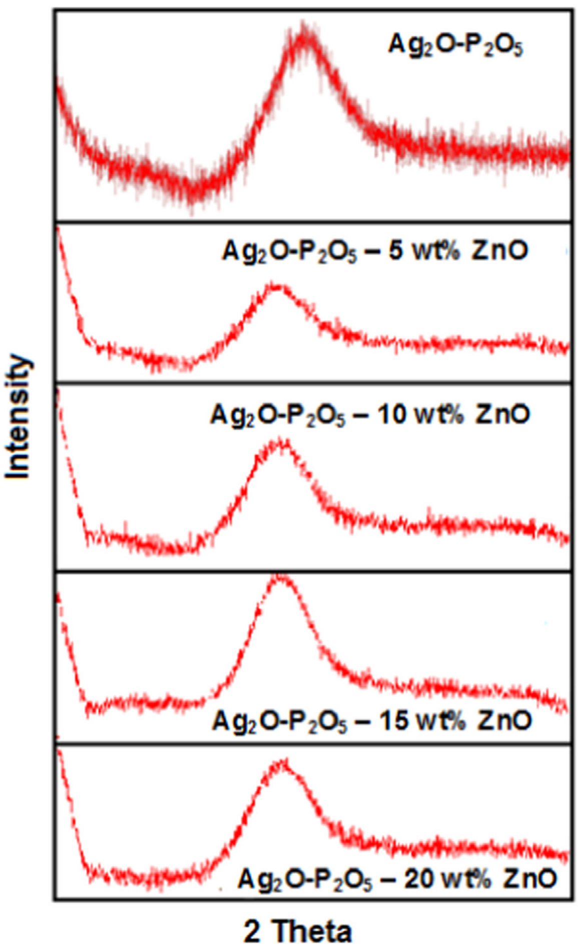

Fig. 1. X-ray diffraction patterns of undoped and doped silver phosphate glasses.

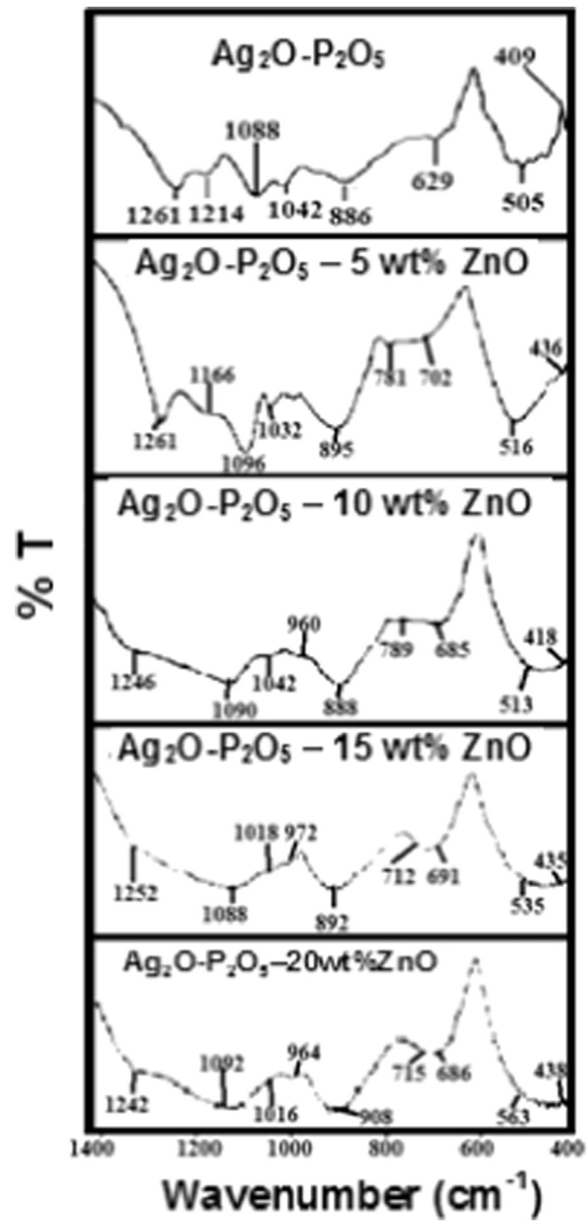

Fig. 2. Infrared spectra of undoped and doped silver phosphate glasses.

2.2.2. Preparation of glasses

Silver phosphate glasses were prepared by melt quench technique [8]. Silver nitrate and ammonium dihydrogen phosphate were mixed thoroughly in 1:1 M ratio in an agate mortar, homogenized and kept in a platinum crucible. The platinum crucible containing the mixture was heated in an oven at 150 °C for about one hour and then in an electric muffle furnace at 800-900 °C for 4 h. The molten mixture was then suddenly quenched to 0 °C to form silver phosphate glass. The process of melting and quenching was repeated three times. In the second phase 5, 10, 15 and 20 wt% nanosize zinc oxide were mixed separately in 1:1 M mixture of silver nitrate and ammonium dihydrogen phosphate and the process was repeated as above. All the glass samples were kept in glass tubes, covered with black cotton and kept in a desiccator.

2.2.3. X-ray diffraction studies

X-ray diffraction patterns of all the glasses were recorded using CuK_{α} radiation.

2.2.4. FT-IR spectral studies

FT-IR spectral studies of all the glasses were made with Shimadzu IR AFFINITY-1 spectrometer at room temperature in the range 2000–400 cm^{-1} using KBr pellets.

2.2.5. SEM studies

In order to know the morphologies of ZnO and the glasses, SEM studies were carried out using Quanta FEG 250 ESEM instrument. The samples were coated with gold.

2.2.6. Differential scanning calorimetric (DSC) studies

In order to determine the glass transition temperatures (T_g), differential scanning calorimetric studies between room temperature to 873 K were carried out with the help of METTLER TOLEDO, DSC 822e instrument. A heating rate of 10 K/min in nitrogen atmosphere was used in this study.

2.2.7. Electrical conductivity and dielectric constant measurements

The electrical conductivities of all the glasses were measured by a.c. impedance spectroscopic method using HIOKI 3532-50 LCR Hi TESTER. Glass pellets were made by applying a pressure of 5 t with the help of a hydraulic press machine. The pellets were coated with silver paste on both the sides and kept between two