UNIVERSITI TEKNOLOGI MARA

STRUCTURAL, ELASTIC AND OPTICAL PROPERTIES OF xSrO-10PbO-(90-x)B_2O_3, xPbO-30SrO-(70-x)B_2O_3 AND xSrO-(90-x)B_2O_3-2CeO_2-8Al_2O_3 BORATE GLASSES IN BORATE ANOMALY REGION

NURUL SYAHIDAH BINTI SABRI

Thesis submitted in fulfilment
of the requirements for the degree of
Doctor of Philosophy

Faculty of Applied Science

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CONFIRMATION BY PANEL OF EXAMINERS

I certify that a Panel of Examiners has met on 25th January 2018 to conduct the final examination of Nurul Syahidah binti Sabri in her **Doctor of Philosophy** thesis entitled "Structural, Elastic and Optical Properties of xSrO-10PbO-(90-x)B_2O_3, xPbO-30SrO-(70-x)B_2O_3 and xSrO-(90-x)B_2O_3-2CeO_2-8Al_2O_3 Borate Glasses in Borate Anomaly Region" in accordance with Universiti Teknologi MARA Act 1976 (Akta 173). The Panel of Examiner recommends that the student be awarded the relevant degree. The Panel of Examiners was as follows:

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ABSTRACT

Glass samples with the composition of xSrO-10PbO-(90-x)B_2O_3 (x = 20-45), xPbO-30SrO-(70-x)B_2O_3 (x = 0-25) and xSrO-(90-x)B_2O_3-2CeO_2-8Al_2O_3 (x = 25-50 mol%) were prepared by melt-quenching method to elucidate the structural, elastic and optical behavior in borate anomaly region. Characterization techniques like X-ray diffraction (XRD), Fourier transforms infra-red spectroscopy (FTIR), ultrasonic measurement, UV–Visible and Luminescence (for SrO-B_2O_3-CeO_2-Al_2O_3) spectroscopes are employed in the present work. XRD data showed amorphous nature for SrO-PbO-B_2O_3 samples while mixed crystalline/amorphous phases for the SrO-B_2O_3-CeO_2-Al_2O_3 samples at x > 35 mol%. FESEM image and EDX analysis revealed that the samples contained Ce-rich crystalline phase and Ce depleted glass phase. FTIR analysis revealed the presence of BO_4 and BO_3 vibration groups. Variation of SrO in SrO-PbO-B_2O_3 and SrO-B_2O_3-CeO_2-Al_2O_3 samples has resulted in increased ultrasonic velocities, elastic moduli (C_L, μ, K and Y), hardness (H), Debye temperature (θ_D) at lower SrO content but decreased for higher SrO addition. The increase in the elastic moduli indicates the increase of network rigidity of the glass system, related to the borate anomaly where the coordination number increased with the addition of SrO. At higher SrO additions in SrO-PbO-B_2O_3 samples, NBO formation resulted in decrease of elastic properties. Meanwhile, decrease in elastic moduli for SrO-B_2O_3-CeO_2-Al_2O_3 samples related to the formation of NBO and grain/phase boundaries due to mixed crystalline/amorphous phases. The fraction of the four coordinated boron atoms (N_4) values, calculated from FTIR spectra have same trend with elastic properties results. Quantitative analysis of ultrasonic data using the bulk compression and ring deformation models showed reduction in the ratio of calculated bulk modulus to the experimental bulk modulus, K_{bc}/K_e indicating decreased ring deformation in borate anomaly region. Observed mismatch in maximum value of optical band gap (E_{opt}) and minimum values of electronic polarizability ($\alpha_{O^{2-}}$), optical basicity (Λ) and refractive index (n) when compared to maximum values of N_4 and elastic behavior was attributed to the formation of weak coordinated covalent bonds during structural transformation and addition of cation with high polarizability to the xSrO-10PbO-(90-x)B_2O_3 glass. Presence of crystalline phases along with the glass matrix in the SrO-B_2O_3-CeO_2-Al_2O_3 glass at x > 35 mol% resulted in higher of E_{opt} and lower of n values. Formation of BO_4 and partial crystallization have resulted in enhancement and quenching of emission spectra in the SrO-B_2O_3-CeO_2-Al_2O_3 glasses, respectively. Meanwhile, by varying PbO content in xPbO-30SrO-(70-x)B_2O_3 samples, elastic moduli, FTIR and optical absorption analysis revealed that PbO acts differently i.e glass former and glass modifier, depending on the PbO concentration. PbO can act as a conditional glass former and its incorporation into glass in the form of PbO_4, which causes considerable change in the structure of glass. In this work, it can be suggested that at $x \leq 10$ mol% of PbO content, Pb^{2+} is incorporated as network modifier, while it acts as network former for the higher PbO content (x > 10 mol%).

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