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Mixed-Alkali Effect in Li_2O–Na_2O–K_2O–B_2O_3 Glasses: Infrared and Optical Absorption Studies

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The mixed-alkali effect (MAE) has been investigated in the glass system $(40 - x)Li_2O$–xNa_2O–$10K_2O$–$50B_2O_3$ (0 mol% ≤ x ≤ 40 mol%) through density, modulated differential scanning calorimetry (DSC), and optical absorption studies. From the absorption studies, the values of the optical band gap (E_{opt}) for direct transition and Urbach energy (ΔE) have been evaluated. The values of E_{opt} and ΔE show nonlinear behavior with the compositional parameter. The density and glass-transition temperature of the present glasses also show nonlinear variation, supporting the existence of MAE. The infrared (IR) spectra of the glasses reveal the presence of three- and four-coordinated boron atoms. The specific vibrations of Li–O, Na–O, and K–O bonds were observed in the present IR study.

Key words: Density, glasses, infrared spectroscopy, optical absorption, band gap energy, Urbach energy, mixed-alkali effect

INTRODUCTION

Nonlinear variation in properties (including density, viscosity, glass-transition temperature, and conductivity) is observed when one alkali component in a multicomponent glass is systematically replaced by another species.[1–3] Besides glasses containing only alkali ions, such nonlinear behavior is exhibited by certain mixed alkali–alkaline earth oxide glasses also.[4] This phenomenon is referred to as the mixed-alkali effect (MAE) and is useful in manufacturing low-loss electrical glass.

Many investigations have been reported on the MAE in phosphate, borate, tellurite, bismuthate, and silicate glasses containing alkali and/or alkaline-earth oxides.[5–12] Raghavender Rao et al.[13,14] reported physical, optical, Fourier-transform infrared (FT-IR), and electron paramagnetic resonance (EPR) studies in mixed-alkali zinc borate glasses doped with nickel and manganese.

Recently, Ganapur et al.[15] presented EPR studies on a mixed-alkali multicomponent glass system. In our previous papers,[16–18] we reported on measurements of density, glass-transition temperature, optical absorption, and EPR of mixed-alkali borate glasses.

Most technical glasses contain a variety of different cations, but almost all studies on the MAE are focused on glasses with only two kinds of ions. Therefore, it is very important to study the ion dynamics of glasses with three or more types of alkali ions. For the first time, Gao and Cramer[19,20] reported the temperature-dependent direct-current (DC) conductivity of a series of mixed-alkali glasses with three types of alkali ions. The results confirmed that the strength of the MAE in conductivity is more pronounced in ternary mixed-alkali borate glasses than in binary mixed-alkali borate glasses.

Kim et al.[21] investigated the temperature- and frequency-dependent ionic conductivity in Li_2O–Na_2O–Rb_2O–B_2O_3 glasses and compared it with that of binary mixed-alkali Li_2O–Rb_2O–B_2O_3 and Li_2O–Na_2O–B_2O_3 and single-alkali Rb_2O–B_2O_3 glasses. They observed that the quaternary system exhibited the combined characteristic of binary mixed-alkali systems, indicating that the dimensionality of the conducting pathway in mixed-alkali glasses is lower

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