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High Temperature Brillouin Scattering of Potassium Borate Glasses

カリウムホウ酸塩ガラスの高温ブリルアン散乱測定

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1. Introduction

The elastic properties of alkali borate binary glasses at room temperature have been investigated systematically [1, 2, 3]. Blow a glass transition temperature (T_g), the physical properties such as density are relatively stable. In contrast, since the rigidity of glass decreases rapidly above T_g, the elastic properties at high temperatures may show marked temperature dependence. Therefore, it is important to understand the elastic properties of alkali borate glasses over a wide temperature range.

It is difficult to measure elastic property of the glass at high temperatures, because the glass became very soft above T_g. In order to measure at even high temperatures, Brillouin scattering can be employed. Brillouin scattering enables to probe the elastic properties in GHz frequency range without any contact with a sample, which is important to study a sample at high temperatures. Thus, Brillouin scattering measurements is a powerful tool for determining the sound velocity and absorption coefficient of glass and glass forming liquid.

In this study, potassium borate glasses $x\mathrm{K_2O}\cdot(100\text{-}x)\mathrm{B_2O_3}$ (x = 4, 10, 14, 20, 28, 34) , where x denotes the molar composition of $\mathrm{K_2O}$, are investigated through the Brillouin scattering up to 1100 °C using a compact IR image furnace.

2. Experimental

All glasses were prepared with high homogeneity in order to investigate the inherent nature of binary system. To achieve the high homogeneity of the glass forming materials, they were made by the reaction in aqueous solutions [2]. The compositions of all glasses were analyzed with respect to both x and (100-x) with a neutralization titration.

The experimental setup of a Brillouin scattering apparatus is described elsewhere [1]. The features of this system are a combination of a microscope and a Sandercock-type 3 + 3 passes tandem multipass Fabry-Perot interferometer (FPI). The Brillouin scattering spectra were measured at a backward scattering geometry. A standard photon counting system and a multichannel analyzer were used to accumulate the signals. The temperature was controlled by the compact IR image furnace (Yonekura, IR-TP) specially customized for the present experiments.

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3. Results and discussion

Figure 1 shows the temperature dependence of Brillouin spectra in $28\mathrm{K_2O}\cdot72\mathrm{B_2O_3}$. The doublet of Brillouin components of the longitudinal acoustic phonon is located on both sides of the center. The Brillouin shift decreases and full width at half maximum (FWHM) increases with increasing temperature above T_g = 400 °C. The appearance of central peaks was confirmed above 700 °C. The longitudinal sound velocity (V_L) and absorption coefficient (α_L) of the samples were calculated from Brillouin shift ($\Delta\nu_{180}$) and FWHM (Γ) using the equations (1) and (2),

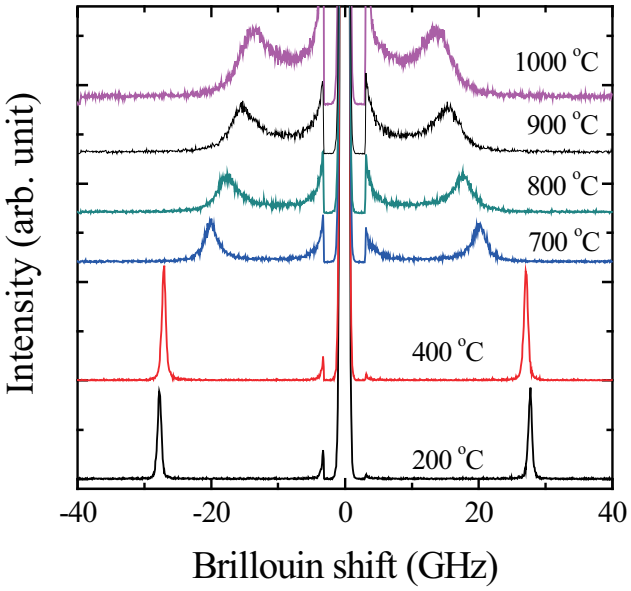

Fig. 1 Temperature dependence of Brillouin spectra in $28\mathrm{K_2O}\cdot72\mathrm{B_2O_3}$.

$$V_L = \frac{\Delta\nu_{180}\lambda}{2n\sin(\theta/2)}, \quad (1)$$

$$\alpha_L = \frac{\pi\Gamma}{V_L}, \quad (2)$$

where n is the refractive index, λ is the wavelength of the incident beam (532 nm) and θ is the scattering angle.

Figures 2 and 3 show temperature dependences of longitudinal sound velocity and absorption coefficient, respectively. At room temperature, V_L increases with increasing K_2O composition. This increase has been discussed by a change of intermediate structure [3]. At temperatures below T_g, V_L decreases gradually with increasing temperature in all compositions. While above T_g, each V_L decrease very rapidly. This inflection point can be identified as T_g. This change in the slope at T_g indicates that the structure of the glass network becomes weaker or softer above T_g. The rate of decrease is higher the higher K_2O composition. The decrease is a direct measure of structure degradation in glass-forming liquid. This rate of degradation can be used as a measure for fragility. Our results indicate that the fragility of potassium borate glasses increases with K_2O composition. The absorption coefficient increases drastically with temperature above T_g while the temperature dependence below T_g is very small and nearly constant. The rate of increase is higher the higher K_2O composition.

The studies by neutron diffraction and Raman scattering [4, 5] revealed that the glass network is disrupted above T_g. As a result, many random connection and non-bridging oxygen, which destroy the borate network and loosely interact with the potassium ion, are formed. On the other hand, the extents of connectivity in glass network remain unchanged from room temperature to T_g. Therefore, above T_g, V_L and absorption coefficient change drastically and below T_g, their changes are very small.

Relaxation modes give rise to the central peak. It is known that the ionic conductivity of alkali borate increases with increasing temperature and alkali composition. Our results show that the intensity of the central peak increases with increasing temperature and K_2O composition. Thus, the behaviors of composition and temperature dependence of the central peak can be correlated with ionic conduction. The central peak may be related to ionic conduction process in the GHz frequency range.

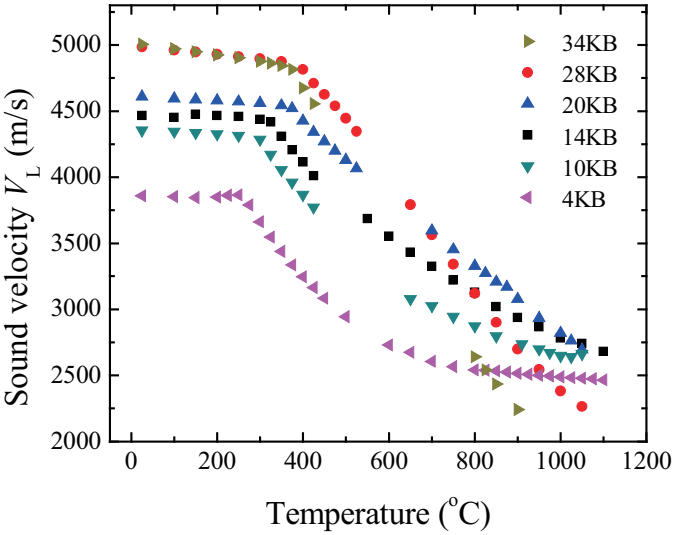

Fig. 2 Longitudinal sound velocity of $xK_2O\cdot(100\text{-}x)B_2O_3$ as a function of the temperature.

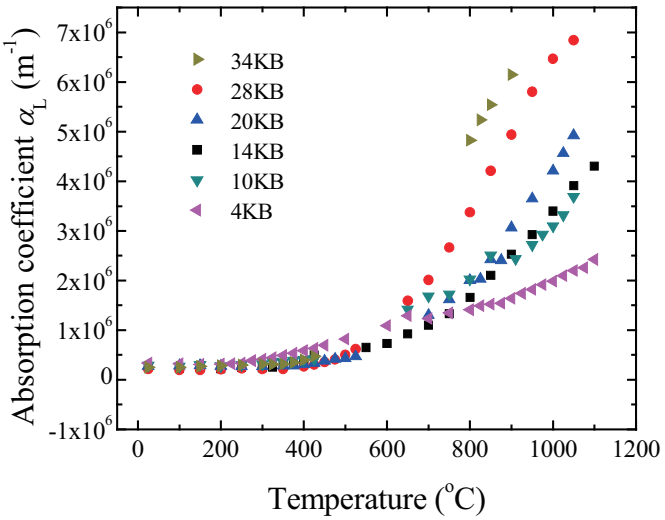

Fig. 3 Absorption coefficient of $xK_2O\cdot(100\text{-}x)B_2O_3$ as a function of the temperature.

Acknowledgment

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