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Internal Friction of $(\text{SiO}_2)_{1-x}(\text{GeO}_2)_x$ Glasses

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Abstract. Internal friction of $(\text{SiO}_2)_{1-x}(\text{GeO}_2)_x$ glasses ($x = 0, 5, 10, 24$ and 100 mole%) is measured at temperatures between 1.6 and 280 K. The data are fitted with the equations for thermally activated relaxation with distributing activation energies in symmetrical double-well potentials. From the determined relaxation strength spectra for each sample, the contributions from each type of microscopic structural units are calculated assuming that transverse motion of the bridging O atom in Si-O-Si, Si-O-Ge or Ge-O-Ge bridge (T model) is the microscopic unit responsible for relaxation. For instance, the magnitude of internal friction for SiO_2 -5% GeO_2 or 24% GeO_2 is calculated from the data of pure SiO_2 , SiO_2 -10% GeO_2 and pure GeO_2 . The calculated results agree well with the measurements of SiO_2 -5% GeO_2 and SiO_2 -24% GeO_2 . Thus T model is shown to be quantitatively consistent with internal friction of SiO_2 - GeO_2 binary glasses.

1. INTRODUCTION

When a component B is introduced into the host glass of initial component A to make a binary glass, how is the internal friction (IF) changed? Several types of change for mixed glasses may be considered. Some doped component modifies the network of the host glass, such as Na_2O in SiO_2 or in GeO_2 . For Na_2O in SiO_2 , a very large increase of IF is observed compared to pure SiO_2 over a wide range of temperature from liquid He region to several hundreds of K[1]. On the other hand, there is suppression of IF for Na_2O in GeO_2 below room temperature, since Na ions may reduce the initial structural units (ISU) which are responsible for the relaxation in pure GeO_2 [2]. If the doped component is the same type of network former as the host glass like GeO_2 in SiO_2 , the magnitude of IF may be given as a superposition of the contributions from the ISU of both pure glasses. In this case, there is possibility of newly generated IF due to new structural units (NSU) generated at the phase boundaries of two components. The magnitude may depend on how well the two components are mixed. In the case of SiO_2 - GeO_2 binary glasses, a network of randomly interconnected RO_4 ($\text{R}=\text{Si}$ or Ge) tetrahedra is realized[3-5].

The origin of a broad internal-friction peak of SiO_2 glass, which has a maximum around 30 to 50 K for the frequencies of kHz to MHz, has been discussed for more than 40 years[6]. From the frequency dependence of the peak temperature T_p , it is considered to be due to thermally activated relaxational motion of a single atom or a small group of atoms in the potential which has more than two minima. There are several simplified microscopic models proposed for the relaxation, such as transverse motion of the bridging O atom across the Si-O-Si bridge (T model)[7], longitudinal motion of O atom along the Si-O-Si bridge (L model)[8] and rotation of SiO_4 tetrahedra by a small angle (R model)[9,10]. There is, however, no general consensus for the most probable model.

On the one hand, in spite of a considerable success of a phenomenological model (tunneling model of two-level system: TLS model) for explaining distinct low-temperature properties in glasses such as specific heat, thermal conductivity below 1K and acoustical properties below 5 K, the actual entity is still not clear in real glasses[11]. The situation is the same for SiO_2 , although strong correlation for the microscopic structural origins between the TLS phenomena and the classical thermally activated relaxation has been suggested[12]. Our study on the structural units for the ultrasonic relaxation in the SiO_2 - GeO_2 binary system could be a key to understand the microscopic origins of the thermal activated relaxation and TLS phenomena. We have claimed that T model is qualitatively preferable to other models to explain our data of SiO_2 -5 mole% GeO_2 and 10 mole% GeO_2 [13]. Here we will show that T model is quantitatively consistent with our new data.

2. EXPERIMENTAL

Since SiO₂-GeO₂ glasses have been well developed because of current applications to the optical waveguides, high quality samples are available from manufacturers. The samples are pure SiO₂, SiO₂-5 mole % GeO₂, 10 mole % GeO₂, 24 mole % GeO₂ and pure GeO₂ glasses which are made by the VAD method, a vapor-phase oxidation process from high purity vapors of SiCl₄ and GeCl₄[14].

Internal friction was determined by resonance methods using a composite oscillator with -18.5° X-cut quartz bar (50 kHz) at temperatures between 1.6 K and room temperature. For relatively low attenuation samples such as pure SiO₂, 5% and 10% GeO₂, the energy required for steady oscillation in a feedback-loop circuit was measured[15]. For high attenuation samples such as 24% GeO₂ and pure GeO₂, the decay of free oscillation was measured using a storage oscilloscope.

The results are shown in Fig.1. The solid lines are fitting results calculated from the equations for thermally activated relaxation as explained below.

3. DISCUSSION

For the analysis of the IF spectra on temperature, we use the Debye equation with angular frequency ω and relaxation time τ for the relaxation in symmetrical double-well potentials (DWP).

$$\Delta = \frac{2\pi ND^2}{EKT} \frac{\omega\tau}{1 + \omega^2\tau^2}, \tag{1}$$

where E is the Young's modulus, D is the deformation potential, N is the number density of the active structural unit for the relaxation and kT has usual meaning. The relaxation time τ for the thermal activation is given as

$$\tau = \tau_0 \exp\left(\frac{V}{kT}\right), \tag{2}$$

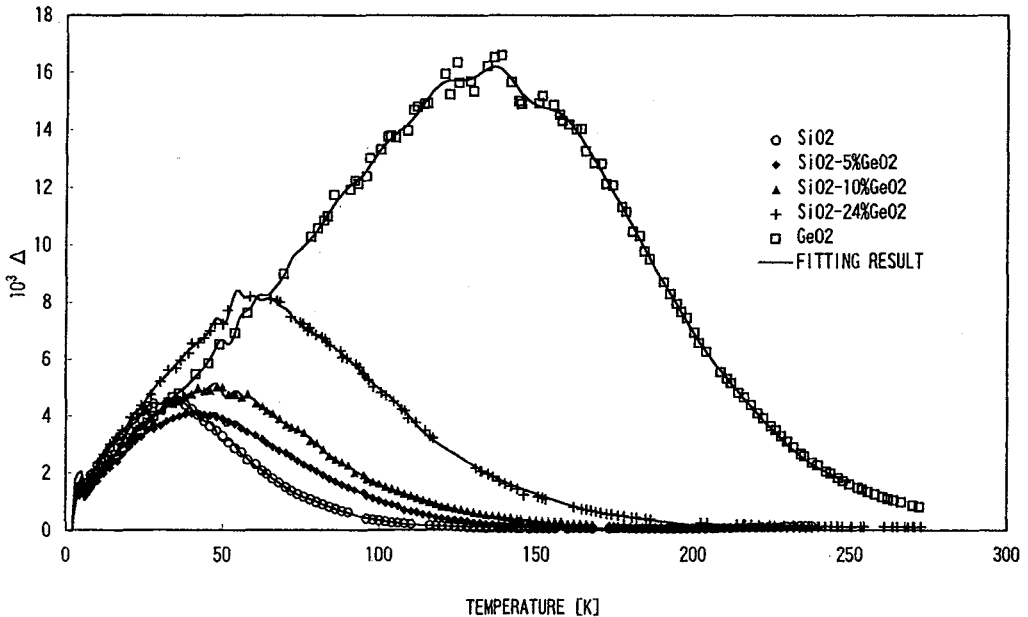
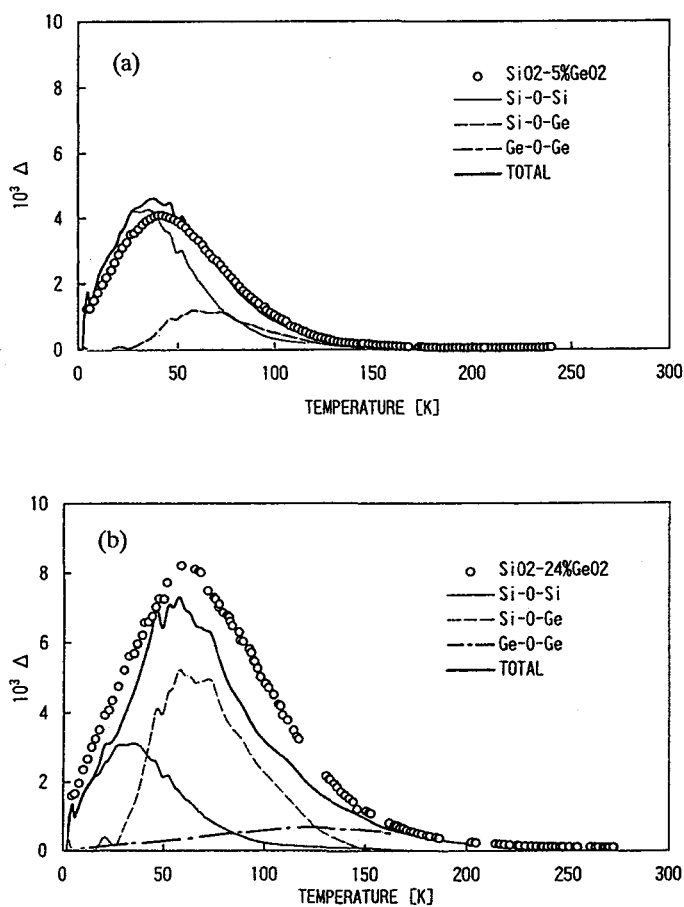


Fig. 1 Internal friction vs temperature at 50 kHz. Solid lines are fitting results of equation (3) to the data.

Table I Fraction of structural units, Si-O-Si, Si-O-Ge and Ge-O-Ge for SiO₂-5%, 10% and 24% GeO₂.

	SiO ₂ -5%GeO ₂	SiO ₂ -10%GeO ₂	SiO ₂ -24%GeO ₂
Si-O-Si	0.902	0.810	0.578
Si-O-Ge	0.095	0.180	0.365
Ge-O-Ge	0.003	0.010	0.058

Fig. 2 Internal friction of SiO₂-5% GeO₂ (a) and SiO₂-24% GeO₂ (b). Lines are calculations from the data of pure SiO₂, SiO₂-10% GeO₂ and pure GeO₂.

where τ_0 is pre-exponential factor of order 10^{-13} s and V is the activation energy or height of the barrier of the double-well potential. Since the barrier height of DWP in glasses is generally distributed, we use the following function for IF to fit to the data.

$$\Delta = \sum_i \frac{A_i}{T} \frac{\omega \tau_i}{1 + \omega^2 \tau_i^2}, \quad \tau_i = \tau_0 \exp\left(\frac{V_i}{kT}\right) \quad (3)$$

Here a set of A_i 's will be determined by fitting the equation to the data for each sample. Thus the solid lines are fitting results (Fig.1).

Next, by using the determined A_i 's we tested the consistency between a model and the data. Since we have claimed that T model is the most probable for the origin of IF among the proposed microscopic models, we concentrate on showing the quantitative consistency of this model. In pure SiO_2 and GeO_2 , we assume that transverse motion of the O atom in the R-O-R bridge (R = Si or Ge) is initial structural unit (ISU) for the relaxation. In the mixed glasses of SiO_2 -5% GeO_2 , 10% GeO_2 and 24% GeO_2 , the number density of ISU such as Si-O-Si or Ge-O-Ge should be changed. In addition the new structural units (NSU) of the transverse motion of the O atom in the Si-O-Ge bridge will be generated. The expected fraction of each type of units in mixed glasses is shown in Table I, assuming that Ge atoms randomly substitute for the sites of Si atoms. For 5% and 10% GeO_2 , the fraction of the Ge-O-Ge is negligibly small (less than 1 %). So we may determine the IF spectra on temperature due to the NSU, for instance, only from the data of pure SiO_2 and SiO_2 -10% GeO_2 . Then we may calculate the expected spectra for SiO_2 -5% GeO_2 and SiO_2 -24% GeO_2 from the determined IF spectra due to NSU and ISU. We checked consistency of the calculated results and the experimental data (Fig.2(a) and (b)). The calculated spectra can explain the data satisfactorily. In particular, the peak temperature is predicted well and the magnitude of the IF is explained within a difference of 20 %. We note that the absolute magnitude of IF of glasses is difficult to determine uniquely since a different heat treatment will give a fluctuation of it in glasses, as by up to 20% in SiO_2 [16]. So we conclude that T model is consistent with all the data quantitatively. It should be noted that for SiO_2 -24% GeO_2 the loss due to the NSU (Si-O-Ge) is considerably large and the contribution of the ISU (Ge-O-Ge) is not negligible to explain the loss above 100 K.

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