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Brillouin Scattering Study of Glass Transitions in Glucose Aqueous Solution
グルコース水溶液のガラス転移のブリルアン散乱

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1. Introduction

Liquid-glass transition occurs by cooling a viscous liquid fast enough to avoid crystallization. The vitrification of saccharides such as glucose aqueous solution have been regarded as one of the effective way to avoid water crystallization in cells, and known as one of typical cryoprotectants.[1,2)] However, the dynamics of liquid and glassy states of glucose has not yet been fully understood. Therefore, it is important to investigate the glass transition behavior of glucose aqueous solutions.[3)]

Brillouin scattering spectroscopy has been used to study the elastic properties of various liquids and glass transition processes in a high-frequency giga-hertz range and in an extremely wide temperature range. Brillouin scattering is a powerful tool to study dynamical properties of glass transitions.[4,5)] We can determine the sound velocity and attenuation accurately by Brillouin scattering.

In this study, the elastic properties of glucose aqueous solutions were investigated at various concentrations of glucose. Sound velocity and attenuation factor of glucose aqueous solutions were determined from Brillouin scattering spectroscopy. The relaxation process is discussed in relation to the glass transition.

2. Experimental

D-(+)-Glucose of 99% purity was purchased from Fluka and no further purification was done. The refractive index was measured by the prism coupling method (Metricon, 2010/M) at 532 nm index accuracy of ±.0005. The sound velocity was measured by Brillouin scattering. [4)] The light source was a green YAG laser (532 nm, 100 mW). The Brillouin scattering spectra were measured using the Sandercock-type 3 + 3 pass tandem Fabry Perot interferometer (JRS, TFP-1) with finess 100 at the backward scattering geometry. The sample temperature in a heating/cooling stage (LINKAM, HTMS600) was controlled from -175 to 120 °C .

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3. Results and Discussion

3.1 Refractive index

The refractive index of glucose aqueous solutions were measured at 532 nm as a function of glucose content as shown in Fig. 1. The refractive index shows linear dependence with the glucose content x. In lower alcohols the refractive index shows non-linear relation with the alcohol content, because associated clusters are generated by the combination of water and alcohol molecules. Therefore, the linear dependence of glucose solutions suggests the low possibility of the generation of associated clusters between water and glucose molecules.

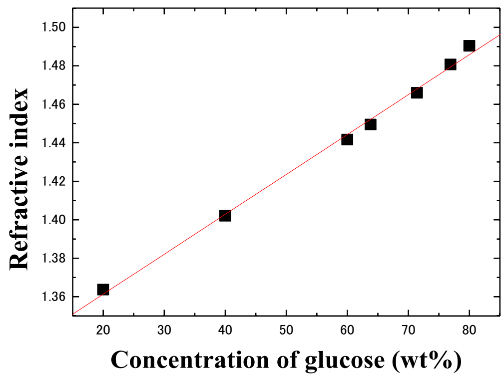

Fig. 1 Refractive index at 532 nm of glucose aqueous solutions at 30 °C as a function of glucose concentration.

3.2 Brillouin scattering

The Brillouin scattering of glucose aqueous solutions were measured as a function of glucose content as shown in Fig. 2. With the increase of the content, frequency shift becomes higher and the width shows broadening. In contrast with the refractive index, the sound velocity shows the non-linear dependence.

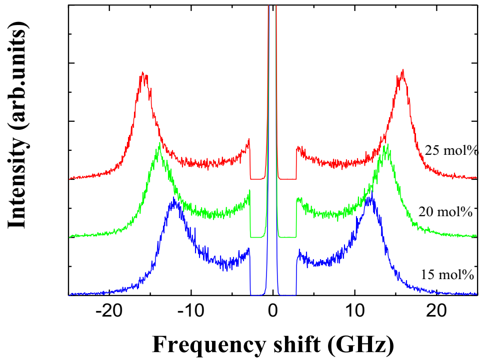

Fig. 2 Brillouin scattering spectra of glucose aqueous solutions at 30 °C as a function of glucose concentration.

The temperature dependence of Brillouin scattering spectra of 40 wt% glucose aqueous solution is shown in Fig. 3. On cooling from 100°C, the broadening of LA peaks is observed by the structural relaxation process related to a glass transition. No intense central peak is observed.

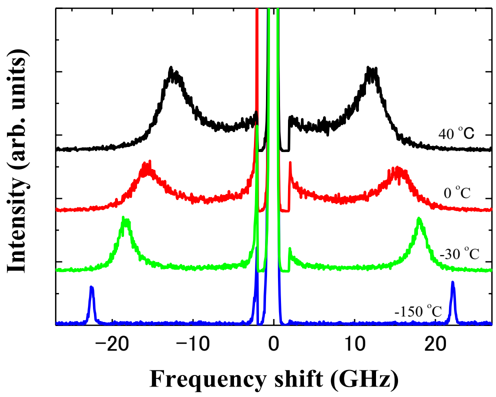

Fig. 3 Temperature dependence of Brillouin scattering spectra of 15 mol% glucose aqueous solution.

Figure 4 shows the temperature dependences of sound velocity and attenuation of the LA mode, which were determined using the observed values of refractive index described in Sec. 3.1. In low glucose contents below 50 wt%, on cooling the crystallization occurs and does not undergo a glass transition for further cooling. While in higher content above 60 wt%, it undergoes a glass transition and shows the change of sound velocity in the vicinity of a glass transition temperature T_g.

As to the structural relaxation above T_g, the peak of attenuation was clearly observed above 50 %. The temperature variations of sound velocity and attenuation were analyzed by the Debye model. The relaxation time of structural relaxation process was determined as a function of temperature. Its temperature dependence shows Arrhenius behavior. The parameters of Arrhenius law were also determined.

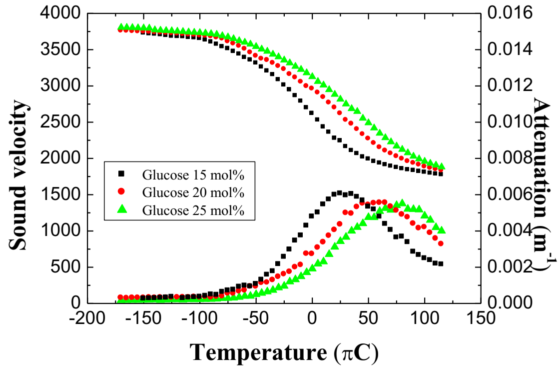

Fig. 4 Temperature dependence of sound velocity and attenuation of the LA mode in glucose aqueous solutions.

4. Conclusion

The low-temperature properties of glucose aqueous solutions has been studied by Brillouin scattering excited at 532 nm. Temperature and composition dependences of refractive index of glucose aqueous solutions have been determined at 532 nm accurately to determine the sound velocity and attenuation. The acoustic anomaly of glucose aqueous solutions shows the typical structural relaxation in the supercooled liquid state, and undergo a liquid-glass transition at low temperatures. The glass transition temperatures were also determined by the change of sound velocity. Our results verify that the glass transition dynamics of glucose aqueous solutions is similar to typical cryoprotectants such as aminopropanol.[6)]

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