

QENS Study on Thermal Gelation in Agarose Aqueous Solution

N. Onoda-Yamamuro(A) and O. Yamamuro(B)
(A) *Tokyo Denki Univ.*, (B) *ISSP-NSL, Univ. Tokyo*

Thermoreversible gelation of aqueous solution of macromolecules is due to the formation of a physically crosslinked network structure. Macroscopic properties, such as viscosity, change drastically with gelation. It is of interest whether microscopic motions such as diffusion of water molecules and segmental vibration of macromolecular chains change or not with gelation. Methylcellulose (MC) gels on heating due to hydrophobic interactions between methoxyl groups. We have shown that dynamic slowing down occurs not only for macroscopic properties but also for microscopic motion of both MC and water molecules by quasi-elastic neutron scattering (QENS) measurements [1]. In this study thermal gelation of agarose was examined. Agarose (AG) gels on cooling due to formation of hydrogen bonds.

QENS experiments were performed on AG solution using AGNES spectrometer, installed at C3-1 port of JRR-3M in Tokai. D₂O, not H₂O, was used as a solvent to observe both dynamics of water and AG molecules. The relative scattering intensity from H atoms of agarose to that from D atoms of D₂O is expected to be comparable in this sample solution. The sample (3.0 wt-% solution) in this study gelled around 315K on cooling and soled around 370K on heating.

The data were fitted well to the sum of a Lorentzian function and a Delta function, both of which are convoluted by resolution function as shown in Fig. 1 (a). The former is attributed to diffusive motion of water molecules and the latter to local vibrational motion of AG molecules. The self-diffusion coefficient D of water molecules was obtained from the Q -dependence of the HWHM of the Lorentzian function, while the mean square displacement $\langle u^2 \rangle$ of AG molecules was obtained from the Q -

dependence of the intensity of the Delta term. Temperature dependence of D and $\langle u^2 \rangle$ are shown in Fig.1 (b). The mean square displacement abruptly decreased and the slope of the self-diffusion coefficient changed with gelation on cooling. The present results revealed that the microscopic motions of both water and AG molecules give rise to dynamic slowing down on thermal gelation.

[1] N. Onoda-Yamamuro et al. *Physica B Condens. Matter*, 393, 158 (2007).

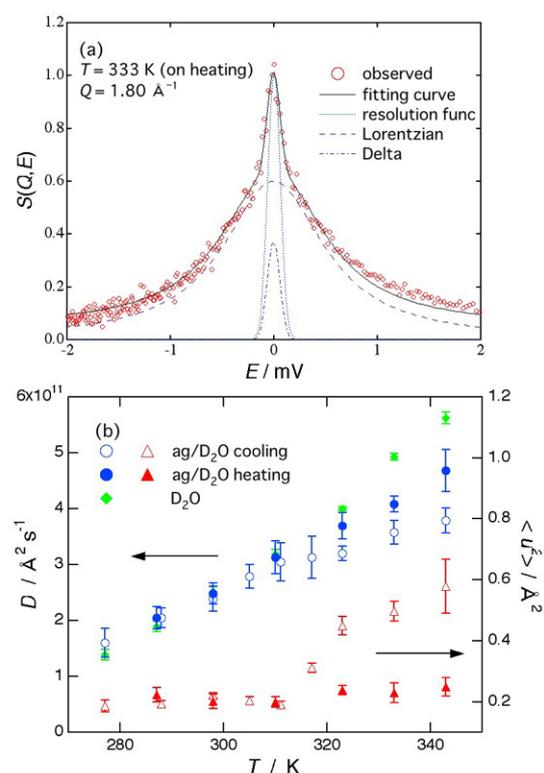


Fig. 1. Figure 1(a) An example of fits to $S(Q,E)$ of the AG solution. (b) T dependence of the self-diffusion coefficient D of water and the mean square displacement $\langle u^2 \rangle$ of the local motion in AG molecules.