Novel critical behavior in a mixture of water / organic solvent under high-pressure condition

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In general, the SANS profiles for water/organic solvent mixtures near the critical point follow Ornstein-Zernike equation, $I = I_0 / (1+\xi^2 q^2)$, where ξ denotes the correlation length of concentration fluctuation, and I_0 the forward scattering. Furthermore, the critical exponents forξ and I_0 represent the values of the 3D-Ising universality (i.e., ν = 0.63 and γ = 1.24). However, we discovered a novel critical behavior that does not follow to the 3D-Ising universality in a water/3-methylpyridine mixture under high pressure. At 298 K, this mixture exhibits phase separation when pressures above 100 MPa are applied. In this case, it was found that the critical exponents show the values of the mean-field (i.e., ν = 0.50 and γ = 1.00), not the 3D-Ising. This result suggests that long-range intermolecular interactions rather than shot-range interactions contribute to phase separation under high pressure.

In this study, we tried to verify whether such a phenomenon is also observed in other kinds of binary mixtures. Unfortunately, just before starting the experiment, it was confirmed that the window material of the high-pressure cell for the SANS experiment had cracked. Therefore, we installed the high-pressure cell at the sample position of SANS-U, and conducted a test experiment under normal pressure only (Fig. 1). With proper alignment, we could observe the critical scattering arising from the water/organic solvent mixtures placed inside the high-pressure cell. Furthermore, it was also confirmed that the assumed observation limit of q-value after repairing the window material is Q > 0.1 Å^{-1}.

Next, we observed the critical scattering of the water/2-butoxyethanol mixtures at atmospheric pressure using a normal sample changer. As shown in Fig. 2, it was confirmed that the profiles are explained by Ornstein-Zernike equation. In the future, we plan to measure the critical scattering of this sample under high pressure conditions.

Fig. 1. A high-pressure cell installed at the SANS sample position for a test experiment.

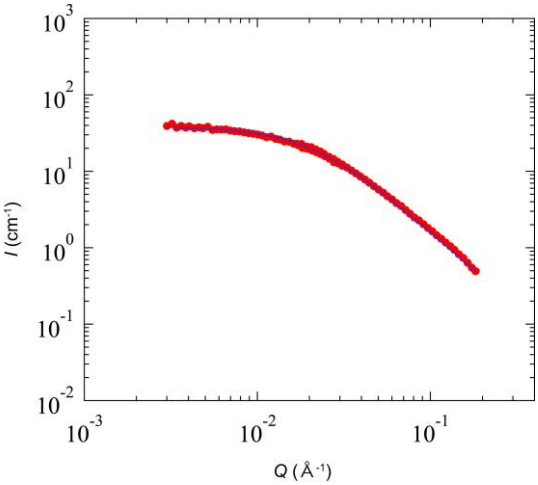

Fig. 2. Critical scattering of water/2-butoxyethanol mixture at 318K. A solid line indicates the fitting result by the Ornstein-Zernike equation.