

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, $I = I_0 / (1 + \zeta^2 q^2)$, where ζ denotes the correlation length of concentration fluctuation, and I_0 the forward scattering. Furthermore, the critical exponents for ζ and I_0 represent the values of the 3D-Ising universality (i.e., $\nu = 0.63$ and $\gamma = 1.24$). 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., $\nu = 0.50$ and $\gamma = 1.00$), not the 3D-Ising. This result suggests that long-range intermolecular interactions rather than short-range interactions contribute to phase separation under high pressure.

Continuing from last year, in the present study, we tried to verify whether such a phenomenon is also observed in other kinds of binary mixtures. Unfortunately, due to two problems, we were not able to carry out the experiment as planned. The first problem is the unidentifiable large background appearing in the low- q region. Figure 1 shows the pressure dependence of the SANS profiles for D₂O/2-butoxyethanol mixture at 310 K. The profile changes with pressure. However, the critical exponent could not be determined due to the large background in the low- q region. The second problem is that the sample is contaminated with surrounding D₂O (pressure medium) during measurement. This problem was caused by the imperfect sealability of the inner cell. The adhesive holding the window in place had leached into the sample during the measurement. As a result, the windows were peeling off (see Fig. 2). This problem could not be solved as long as organic solvents were used for the samples. Therefore,

in the next experiment, it is necessary to use a method of fixing the window material without using an adhesive.

In this manner, we need to address the large background problem shown in the low- q region and the inner cell sealing problem. After overcoming these problems, I would like to repeat the experiment in the future.

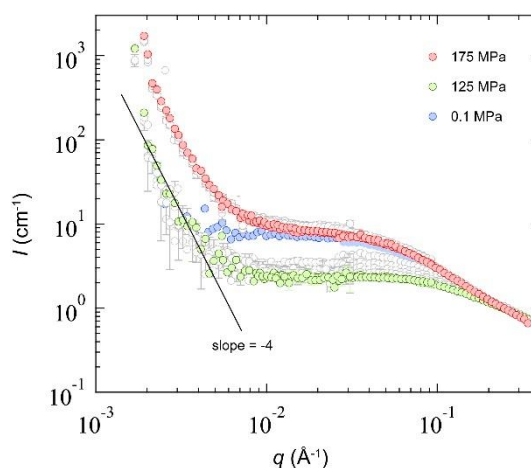


Figure 1: Pressure dependence of the SANS profiles for D₂O/2-butoxyethanol mixture at 310 K. The large unverified background (slope = -4) is shown in the low- q region.



Figure 2: Photograph of the inner cell immediately after the pressure experiment.