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

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Typically, SANS profiles for water/organic solvent mixtures near the critical point adhere to the Ornstein-Zernike equation, $I = I_0 / (1+\xi^2 q^2)$, where ξ represents the concentration fluctuation's correlation length, and I_0 denotes forward scattering. Additionally, the critical exponents for ξ and I_0 align with the 3D-Ising universality values (ν = 0.63 and γ = 1.24). However, we observed an unusual critical behavior in a water/3-methylpyridine mixture under high pressure, which deviates from 3D-Ising universality. This mixture undergoes phase separation at 298 K when subjected to pressures exceeding 100 MPa. In this scenario, the critical exponents were found to match the mean-field values (ν = 0.50 and γ = 1.00) rather than those of 3D-Ising. This finding indicates that long-range intermolecular interactions, rather than short-range interactions, play a significant role in phase separation under high-pressure conditions.

Last year, owing to a defect in the inner cell of the high-pressure cell, the sample leaked outside and correct measurement results could not be obtained. Therefore, in this study, we conducted the experiment again using an improved quartz inner cell, as shown in Figure 1. As a result, we were able to perform measurements with the sample confined inside the inner cell up to the high-pressure region. However, the thickness of the inner cell (4 mm) was too large and the neutron transmittance of the sample was insufficient.

Therefore, sample-derived scattering was not correctly measured. Therefore, in the next experiment, we plan to design a new inner cell with a thinner inner cell thickness of 1 or 2 mm and perform the measurement again.

In addition to experiments under high-pressure conditions, we also measured the dynamics of concentration fluctuations in water/organic solvents using the iNSE in 2023; Research title is “2D-Ising like critical behavior in mixtures of water/organic solvent/antagonistic salt”. Since this was only a preliminary experiment to confirm that a sample-derived signal could be obtained, we plan to conduct the experiment again in 2024.

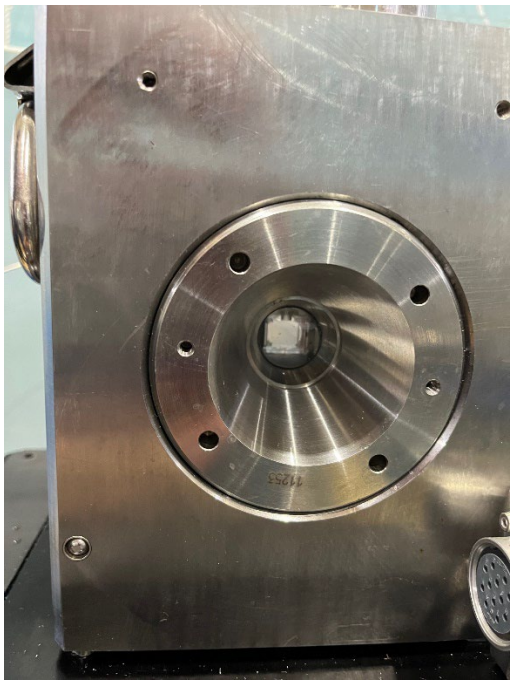

Figure 1: Photographs of the high-pressure cell (stainless steel part) and inner cell (inner part of the central window material); in the 2022 experiment, the sample leaked outward due to a defect in the inner cell, but in the 2023 experiment, as shown in this figure, the sample could be stably sealed up to high-pressure conditions.