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Neutron Spin Echo Studies on Effects of Temperature and Pressure on Structural Phase Transitions and Dynamics in Complex Fluids involving amphiphiles

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In an amphiphilic complex fluid composed of water, oil and amphiphiles, water and oil are separated from each other by amphiphilic membranes. Amphiphiles self-assemble various mesoscopic structures of either disordered or ordered type, such as bicontinuous or lamellar structures. The structural phase transitions are induced by temperature, pressure, composition, salinity and so on. Small angle X-Ray and neutron scattering (SAXS, SANS) are the most important technique to study the static structures of amphiphilic fluids of mesoscopic scales. Neutron spin echo (NSE) is an unique technique to elucidate dynamics of the mesoscopic structure in amphiphilic fluids.

In this study, the mesoscopic structure and its dynamics in the two surfactant systems were investigated by means of SANS and NSE in order to clarify the mechanism of the structural phase transitions induced by temperature and pressure in amphiphilic fluids. I improved the NSE spectrometer and developed a high pressure cell for the NSE experiments.

Dynamics in the nonionic surfactant $C_{12}E_5$/water/*n*-octane system was investigated using NSE in order to clarify the mechanisms of the phase transition from a low temperature microemulsion phase to a high temperature microemulsion phase via a lamellar phase with increasing temperature. The intermediated correlation function $I(Q, t)$ obtained from the NSE experiments were well fitted to a stretched exponential function $I(Q, t) = I(Q, 0)\ \exp[-(\Gamma t)^{2/3}]$. The NSE results supported the theory proposed by Zilman and Granek to describe membrane undulations in a sponge or lamellar phase. Using the theory, we estimated the bending modulus κ of the amphiphilic membranes, which plays an important role in structural phase transitions. κ decreased monotonously with increasing temperature. I conclude that the theory is useful to describe dynamics and analyze the NSE data in a bicontinuous or lamellar phase.

In the ionic surfactant Aerosol OT/water/*n*-decane system, it has reported that the same structures are induced by increasing either temperature or pressure. In order to verify the effects of temperature and pressure, the SANS and NSE experiments were performed in a dilute droplet phase. The NSE results were analyzed with the theory proposed by Milner and Safran, in which the shape deformation of droplets was taken into account. The temperature and pressure dependencies of κ could be obtained on the basis of bending free energy of monolayers without any assumption for the first time. With increasing temperature, the membrane became flexible. On the other hand, it became rigid with increasing pressure. This result confirmed that the effects of temperature were different from those of pressure although the same structures were induced by either temperature or pressure. I conclude that temperature affects mainly the head-head interactions of AOT molecules, while pressure affects mainly the tail of AOT molecules in the

mechanism of the phase transitions.

Key words: neutron spin echo, amphiphilic fluids, slow dynamics, pressure, structural phase transition