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Re-Entrant Direct Hexagonal Phases in a Lyotropic System Induced by Ionic Liquids

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Abstract : The most well-known structures of lyotropic liquid crystalline systems are the two dimensional hexagonal phase of cylindrical micelles with a positive interfacial curvature and the lamellar phase of flat bilayers with zero interfacial curvature. In aqueous solution of surfactants, the concentration dependent phase transitions have been investigated extensively. However, instead of changing the surfactant concentrations, the local curvature of an aggregate can be altered by tuning the electrostatic interactions among the constituent molecules. Intermediate phases with non-uniform interfacial curvature are still unexplored steps to understand the route of phase transition from hexagonal to lamellar. Understanding such structural evolution in lyotropic liquid crystalline systems is important as it decides the complex rheological behavior of the system, which is one of the main interests of the soft matter industry. Sodium dodecyl sulfate (SDS) is an anionic surfactant and can be considered as a unique system to tune the electrostatics by cationic additives. In present study, imidazolium-based ionic liquids (ILs) with different number of carbon atoms in their single hydrocarbon chain were used as the additive in the aqueous solution of SDS. At a fixed concentration of total non-aqueous components (SDS and IL), the molar ratio of these components was changed, which effectively altered the electrostatic interactions between the SDS molecules. As a result, the local curvature is observed to modify, and correspondingly, the structure of the hexagonal liquid crystalline phases are transformed into other phases. Polarizing optical microscopy of SDS and imidazole-based-IL systems have exhibited different textures of the liquid crystalline phases as a function of increasing concentration of the ILs. The small angle synchrotron x-ray diffraction (SAXD) study has indicated the hexagonal phase of direct cylindrical micelles to transform to a rectangular phase at the presence of short (two hydrocarbons) chain IL. However, the hexagonal phase is transformed to a lamellar phase at the presence of long (ten hydrocarbons) chain IL. Interestingly, at the presence of a medium (four hydrocarbons) chain IL, the hexagonal phase is transformed to another hexagonal phase of direct cylindrical micelles through the lamellar phase. To the best of our knowledge, such a phase sequence has not been reported earlier. Even though the small angle x-ray diffraction study has revealed the lattice parameters of these phases to be similar to each other, their rheological behavior has been distinctly different. These rheological studies have shed lights on how these phases differ in their viscoelastic behavior. Finally, the packing parameters, calculated for these phases based on the geometry of the aggregates, have explained the formation of the self-assembled aggregates.

Keywords : lyotropic liquid crystals, polarizing optical microscopy, rheology, surfactants, small angle x-ray diffraction
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