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Phase Transition Behavior of Smectic Phases in Binary Mixture of Liquid Crystals

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The binary mixture of two nonmesogenic compounds viz., Didodecyl dimethyl ammonium bromide (DDAB) and ethylene glycol (EG) exhibits very interesting liquid crystalline phase's at large range of concentrations and temperature. The concentrations with lower/higher percentage of DDAB exhibit I-SmA-SmC-SmE-K sequentially when the specimen is cooled from isotropic phase. Different liquid crystalline phases observed in the mixture were studied using DSC, X-ray, and Optical microscopic techniques. The temperature variations of optical anisotropy have also been discussed.*

Keywords Binary mixture; nonmesogenic; optical studies; phase transition; smectic

Introduction

Thermotropic and lyotropic liquid crystals are the materials, which possess one or more mesophases between the isotropic liquid and the crystalline solid. The sequence of mesophases observed when the temperature is changed, is due to a sequential ordering of molecular arrangements. The shape of molecules is fundamental to determine the features of this sequence. For rod-like molecules, the material can achieve a nematic phase characterized by an orientational order of molecules. In smectic phases, the molecular order increases besides the orientational order, the molecules have an arrangement in layered structure, in some case the molecules are tilt or show positional order within the layers [1–4]. Frequently micellar nematic phase is encountered in the mixture of and glacial acetic acid. It has been also demonstrated that a micellar nematic to cholesteric transition may be induced by addition of optically active compounds [5,6].

In the present investigation, we have shown the existence of lamellar smectic-A, smectic-E, and Smectic-B phases in binary mixture of Didodecyl dimethyl ammonium bromide (DDAB) and Ethylene glycol (EG). The polymorphic smectic modification exhibited on the basis of the results obtained from DSC, X-ray optical anisotropy, and birefringence studies.

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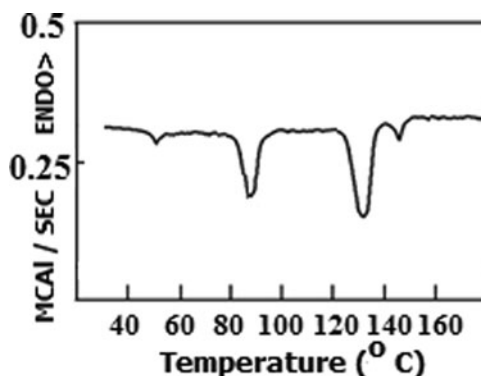


Figure 1. DSC thermogram for the samples of 30% of DDAB in EG.

Experimental Section

The compound Didodecyl dimethyl ammonium bromide (DDAB) used in this investigation was obtained from the Basic Pharma Life Science Pvt., Ltd., India, and it was further purified twice by a re-crystallization method using benzene as a solvent. Ethylene glycol (EG) was supplied from Kodak, Ltd., Kodak house, Mumbai, India. Mixtures of different concentrations of DDAB in EG were prepared and were mixed thoroughly and these mixtures were kept in desiccators for a long time. The samples were subjected to several cycles of heating, stirring, and centrifuging to ensure homogeneity. The phase transition temperatures of these concentrations were measured with the help of Leitz-polarizing microscope in conjunction with a hot stage. The samples were sandwiched between the slide and cover slip and were sealed for microscopic observations. The differential scanning calorimetry (DSC) thermograms were taken for the mixtures of all concentrations using Perkin-Elmer DSC II Instrument facility available at Raman Research Institute, Bangalore, India. The DSC thermogram for the sample of 30% of DDAB in EG is shown in Fig. 1. The X-ray broadening peaks were obtained at different temperatures using JEOL diffractometer. The density and refractive indices in the optical region are determined at different temperatures by employing the techniques described by the earlier investigators [7,8].

Results and Discussion

Phase Diagram

Mixture of DDAB in EG exhibits very interesting different liquid crystalline phases and the phase transition temperatures are measured by using Leitz-polarizing microscopic. The partial phase diagram is shown in Figure 2, and it is obtained by plotting the concentrations against the phase transition temperatures of the mixture, which clearly illustrates that the mixture of all concentrations of DDAB in EG exhibits an induced smectic phases, such as SmA, SmC*, and SmE phases, respectively, at different temperatures, when the specimen is cooled from its isotropic liquid phase. But in concentration range from 5%–27% of DDAB show an unusual chiral SmC* phase increase and hence decrease from 27%–42% of DDAB. The range from 5% to 50% of DDAB shows SmE phase, which remains up to room temperature.

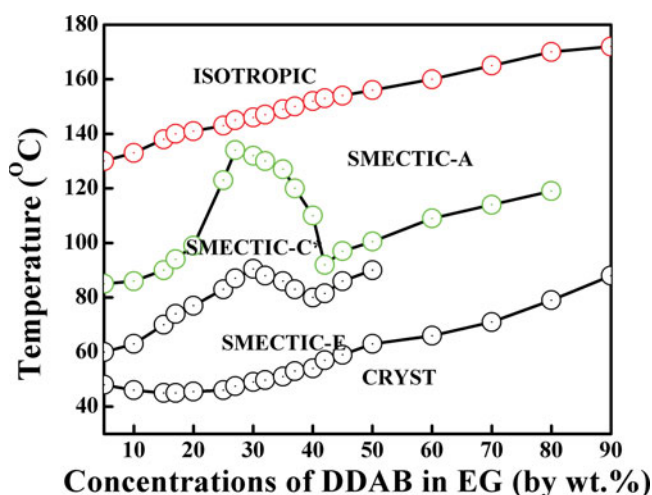


Figure 2. Partial phase diagram for the mixture of DDAB in EG.

Optical Texture Studies

For the purpose of optical texture studies, the sample was sandwiched between the slide and cover glass, and then the optical textures were observed using Leitz-polarizing microscope in conjunction with specially constructed hot stage. The concentrations of the mixture ranges from 5% to 90% are slowly cooled from its isotropic melt, the genesis of nucleation at several points which appear as minute bubbles initially, but which progressively grow radially and form a focal conic fan texture of smectic-A phase in which the molecules are arranged in layers and the texture is shown in Figure 3(a). On further cooling the specimen, this phase appears to be metastable and undergoes slow transformations to give a SmC^* phase, which exhibits a radial fringes on the fans of focal conic textures, these are the characteristics of chiral SmC^* phase as shown in Fig. 3(b). On further cooling the specimen, the SmC^* phase changes over to SmE phase as shown in Fig. 3(c), and then which remains up to room temperature, and then it becomes a crystalline phase.

Birefringence Studies

Results of this investigation are further supported by the optical studies. The refractive indices for extraordinary ray (n_e) and ordinary ray (n_o) of the mixture were measured



Figure 3. Microphotographs obtained in between the crossed polars, (a) Focal conic fan shaped texture of SmA (Lamellar) phase (250 \times). (b) Chiral texture of SmC^* phase (250 \times). (c) Focal conic fans with radial striation of smectic-E phase (250 \times).

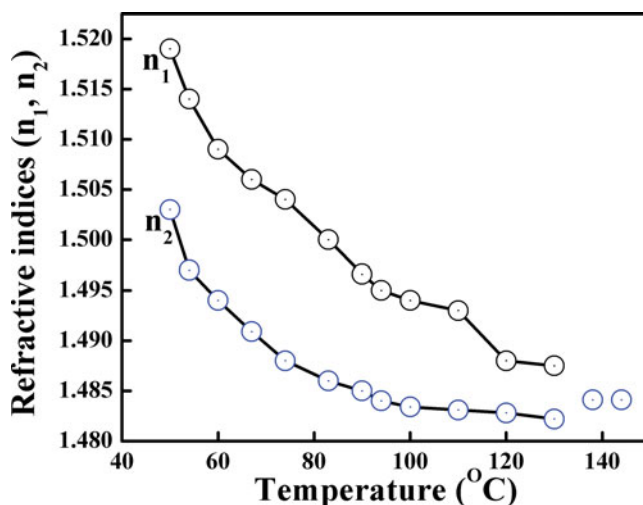


Figure 4. Temperature variations of refractive indices for the mixture of 30% DDAB in EG.

at different temperatures for the different concentrations using Abbe Refractometer and Precision Goniometer Spectrometer. The variations of refractive indices as a function of temperature for 30% of DDAB in EG are shown in Fig. 4. The value of n_e is greater than n_o , indicating that the material is uniaxially positive. The values of electrical susceptibility for 30% of DDAB in EG have been calculated using Neugebauer relation [9] at different temperatures. The variation of electrical susceptibility as a function of temperature for the mixture is shown in Fig. 5. From the figure, it can be observed that wherever there is an isotropic–liquid crystalline phase transition, the value of electrical susceptibility changes appreciably, which indicates that the changes correspond to various smectic modifications. Further, with increase in the concentration of DDAB, the value of electrical susceptibility

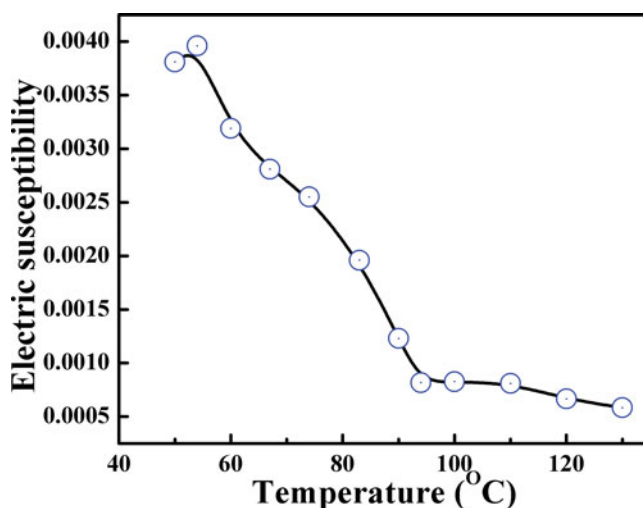


Figure 5. Temperature variation of electric susceptibility for the mixture of 30% DDAB in EG.

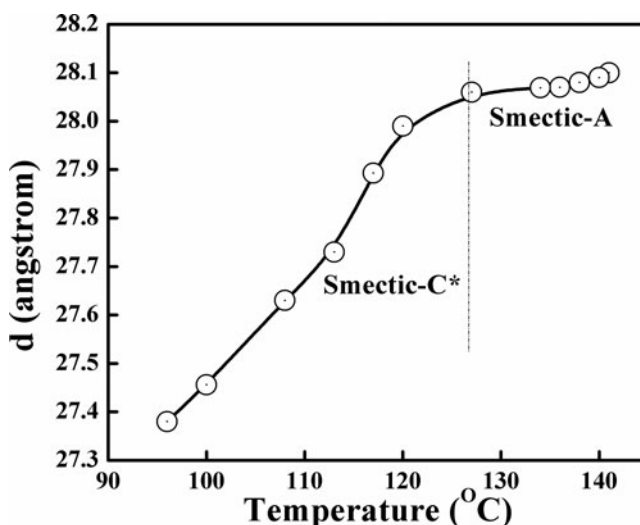


Figure 6. Variation of layer spacing with temperature for the sample of 30% DDAB in EG.

decreases with temperature because the effective optical anisotropy associated with the molecules of DDAB also decreases.

X-Ray Studies

To understand the change in layer spacings in SmA and SmC* phases with respect to temperature, X-ray diffractometer traces were taken. The traces obtained for the mixture of 30% DDAB in EG at different temperature correspond to SmA and SmC* phases. It is observed that as the temperature increases, the layer spacing also increases in SmC* phase, but in SmA phase, the layer spacings are almost constant, and these variations are shown in Fig. 6 [10–12].

Conclusions

Microscopic investigation of the binary mixture of DDAB and EG shows the existence of SmA, SmC*, and SmE phases for all concentrations of DDAB. The phase behavior is discussed with the help of phase diagram. The drastic changes in the values of density, refractive index, and anisotropy of polarizability with the variation of temperature unambiguously correspond to polymorphic smectic phases, respectively. X-ray and DSC studies also lend support to these observations.

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