

Formation of Iridescent Solutions of Dimethylalkylamine Oxides

Aqueous solutions of dimethylalkylamine oxides (C_n DAO, $n = 16, 18$) present the iridescence at surfactant concentrations of 0.3–2 wt%, when the temperature of solutions is lower than 23°C for C_{16} DAO and 46°C for C_{18} DAO. The color changes from yellowish red to blue with an increase in surfactant concentration, and disappears at pH below ca. 6.5 for C_{16} DAO and below ca. 4.5 for C_{18} DAO. For aqueous solutions of C_{18} DAO below ca. 50°C, the weak birefringence occurs at surfactant concentrations above 2.5 wt%, while the bright birefringent color is observed above 9 wt%. © 1989 Academic Press, Inc.

It has been reported that aqueous solutions of polyoxyethylene oleyl ether mixed with a small amount of sodium dedecyl sulfate (1) and aqueous solutions of diglycerol alkyl ethers (2) and alkenylsuccinic acids (3) present the iridescent color at dilute surfactant concentrations of 1–4%. The iridescence phenomenon was also observed in aqueous solutions of dimethyltetradecylamine oxide, when some cosurfactant and a small amount of hydrocarbon were added (4). Color changed with increasing the surfactant concentration. It was suggested that the iridescence was induced by the formation of vesicles with the multilamellar structure (1) or of the multilamellar liquid crystal (2–4).

We have investigated aqueous solutions of dimethylalkylamine oxides (C_n DAO, $n = 16, 18$) and found that they present the iridescence at very dilute surfactant concentrations. The appearance of iridescence originating in the anisotropy of the solution is examined in connection with the occurrence of birefringent color indicating the optical anisotropy.

C_n DAO ($n = 14, 16$) were the same samples as previously prepared and used (5). A sample of C_{18} DAO was kindly given as a gift by Drs. U. Nishimoto and K. Hattori of Kao Corporation, Inc., Wakayama, and recrystallized from an ethanol–acetone mixture. After the surfactant was dissolved in water at high temperature, the solution was cooled and kept 3 h or more at a desired temperature. The surfactant concentration, c , is represented by g/100 g solution (wt%).

The occurrence of iridescence was visually inspected in a glass tube. The inspection was carried out at several temperatures in a water bath. The appearance of birefringent color was observed on an Olympus BH microscope under the crossed nicol. A solution was put in a cell of 1.5 mm depth with glass windows and was kept in a cell chamber with a temperature jacket on the stage of a microscope. The temperature of a cell was measured by a surface thermometer.

Figures 1 and 2 show the region of temperature and surfactant concentration where iridescent solutions of C_{16} DAO and C_{18} DAO are obtained. C_{14} DAO gives only

transparent and isotropic solutions without any iridescence at temperatures down to 0°C and at surfactant concentrations up to 3 wt%.

Aqueous solutions of C_{16} DAO display the iridescence at temperatures below 23°C and at surfactant concentrations above 0.3 wt%, while solutions below 0.3 wt% are only slightly turbid. Color of the solutions changes with their concentrations: red and yellow at 0.5 and 0.7 wt%, yellowish green at 1 wt%, and blue at 1.2–1.9 wt%. The solutions above 1.9 wt% lose the color and are slightly turbid. While color of the solutions does not change within temperatures up to 23°C, it disappears above 23°C and the solutions become transparent and isotropic above 30°C through the turbid regions between 23° and 30°C.

Although aqueous solutions of C_{18} DAO give an iridescence between 0.3 and 2.2 wt%, similar to those of C_{16} DAO, the temperature region of iridescent solutions of C_{18} DAO is below 46°C and is higher than that of C_{16} DAO. Aqueous solutions of C_{18} DAO are transparent and isotropic above 49°C.

The iridescence of aqueous solutions of C_{16} DAO and C_{18} DAO becomes weak upon adding HCl and is not appreciable at pH below ca. 6.5 for C_{16} DAO and below ca. 4.5 for C_{18} DAO. Moreover, the iridescence disappears even in the addition of a small amount of NaOH.

Figure 2 includes the phase diagram of the appearance of birefringence in aqueous solutions of C_{18} DAO. Blue patches are observed for aqueous solutions below ca. 50°C and at 2.5–9 wt%, while bright and colorful birefringent mosaics are observed above 9 wt%. The maximum temperature for the appearance of birefringence is equivalent to the transition temperature from turbid solutions to transparent ones.

The iridescence phenomenon for aqueous surfactant solutions was explained as a result of the interference of light arising from the Bragg reflection (1–4). That is, surfactant molecules associate together to form multilamellar layers in iridescent solutions. The lamellar bilayers can contain a large amount of water between them in dilute surfactant solutions. When the separation between lamellar bilayers is comparable to a half of a wavelength of visible

light, the interference of light reflected from two adjacent bilayers becomes visually appreciable: iridescence becomes visible.

The interpretation above may be applicable to aqueous solutions of C_{16} DAO and C_{18} DAO. However, it should be noted that the range of concentrations of C_{18} DAO for optically anisotropic solutions does not necessarily coincide with that for iridescent solutions but it is located at higher concentrations. This observation on C_{18} DAO is different from that on the iridescent solutions previously investigated (1-3). Their iridescence was observed at surfactant concentrations where the birefringence due to the formation of the liquid crystal occurred.

Since the anisotropic lamellar liquid crystal structure formed by diglycerol alkyl ethers and alkenylsuccinic acids arranges remarkably even in dilute concentrations, the dilute solutions are birefringent and simultaneously iridescent. Similarly, the anisotropic structure constituted by molecules of C_{16} DAO and C_{18} DAO is formed above 0.3 wt%, and the iridescence is induced between 0.3 and 2 wt%. However, the structure is too loose to induce the birefringence at surfactant concentrations below 2.5 wt%, but is more tight between 2.5 and 9 wt% and most tight above 9 wt%.

The solutions of C_{16} DAO and C_{18} DAO were iridescent when the solutions once heated were cooled down below the Krafft point, different from the iridescent solutions previously investigated which were observed above the

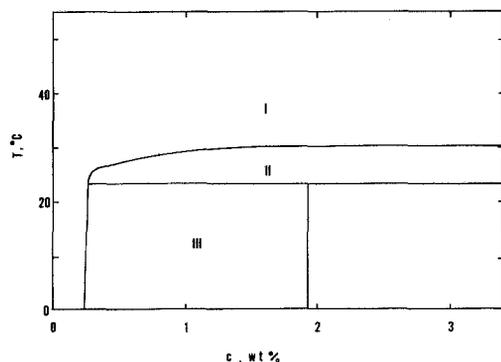


FIG. 1. The phase diagram under the visual inspection for aqueous solutions of C_{16} DAO. I, transparent and isotropic solution; II, turbid solution; III, iridescent solution.

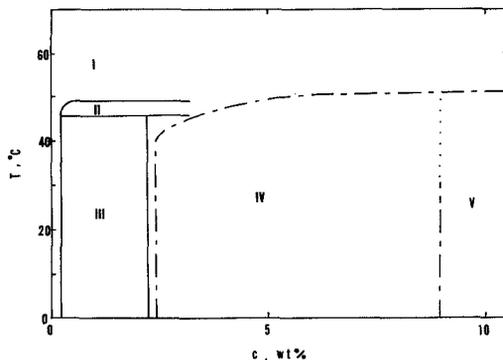


FIG. 2. The phase diagram under the visual inspection and the crossed nicol for aqueous solutions of C_{18} DAO. —, visual inspection; - - -, crossed nicol. I, transparent and isotropic solution; II, turbid solution; III, iridescent solution; IV, weakly birefringent solution; V, strongly birefringent solution.

Krafft point (1-3). This also may indicate the difference of the anisotropic structures between them.

REFERENCES

1. Nagai, M., and Onishi, M., *J. Soc. Cosmet. Chem. Japan* **18**, 19 (1984).
2. Suzuki, Y., and Tsutsumi, H., *Yukagaku* **33**, 786 (1984).
3. Satoh, N., and Tsujii, K., *J. Phys. Chem.* **91**, 6629 (1987).
4. Thunig, C., Hoffmann, H., and Platz, G., *Progr. Colloid Polymer Sci.*, in press.
5. Abe, A., Imae, T., Shibuya, A., and Ikeda, S., *J. Surf. Sci. Technol.* **4**, 67 (1988).

TOYOKO IMAE¹
MOTOI SASAKI
SHOICHI IKEDA

Department of Chemistry
Faculty of Science
Nagoya University
Chikusa, Nagoya 464, Japan

Received April 7, 1989

¹ To whom correspondence should be addressed.