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BILAYER STRUCTURES OF ARTIFICIAL LIPIDS IN A WATER-SOLUBLE POLYMER BINDER

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Artificial ammonium type lipids form bilayer structures in water-soluble polymer film similar to those in water dispersion. Bilayer structures were also formed even when the content of the hydrophobic substance is considerably high (50 mole%).

Various artificial lipids have been synthesized and their aggregate structures in water dispersion have been studied.[1,2,3] The authors are interested in the aggregate structures of lipids in a polymer binder from the viewpoint of the application of lipid functions in solid state. This report describes the observed bilayer structures of artificial lipids in polymer binder.

Commercial poly(vinyl alcohol) (n≑1,750) was used as the water-soluble polymer binder. Didodecyl-, dioctadecyl- and didococyldimethylammonium bromide (**1**,**2**,**3**, respectively) were synthesized by the method proposed by Kunitake et al.[1] N-(γ-morpholinopropyl)-2-hydroxy-3-naphthamide (**4**) was commercially available. Stock solutions for the electron microscopic observation were prepared by dispersing 320 mmole of ammonium salts in 5 ml of an aq.PVA solution. To this solution was added an aq.uranyl acetate solution for aggregate staining and was further diluted in order to obtain a suitable thickness on the Cu-mesh after drying. This solution was applied on a Cu-mesh (150 mesh) both with and without 200Å-thick carbon film. A thin film was formed between the grid. An electron microscope (Hitachi H-700H) in the transmission mode was used for the observation.

Figure 1(a) is an electron micrograph of PVA film containing **1**. Lamellar structures with a width of ca.40Å was observed. This width of 40Å is approximately the same as that obtained by assuming a bilayer structure.[4] It was observed that **1** shows no bilayer structure when 20 mole% (1.9 wt%) of PVA was added to the aq. dispersion of **1**.[5] Distinct bilayer structures, however, were observed in this study when a much larger amount (more than 1700 mole%, 60-100 wt%) of PVA necessary to form a film was added. Similarly, **2** and **3** were found to form bilayer structures in the PVA binder (Figs.1(b) and 1(c)). Not only lamellae but also vesicles are observed in those figures. This suggests that these structures were derived from the aggregate which already existed in the aq.PVA dispersion before drying, and not the microcrystals formed during the drying process. In the case of **2**, lamellar structures were observed in water dispersion.[1] A number of vesicles are shown in Fig.1(b). This is probably due to polymer matrix effect. Hydrophobic substance 4 does not form aggregate by itself in water but it is easily incorporated in the 1-aggregate. Figure 1(d) is an electron micrograph of film obtained from the dispersion containing PVA, 1 and 4 (50 mole%). Multilayer vesicles with a width of ca. 30-40Å were observed. It is noteworthy that bilayer structures were formed in the polymer binder even when the content of the incorporated substance was considerably high.

Thus, it was confirmed that bilayer structures of artificial lipids were formed in polymer binder such as water-soluble polymer here, as well as hydrophobic polymer[4], in spite of the incorporated substance. Proper selection of these polymers will make it possible to utilize lipid functions in solid state.

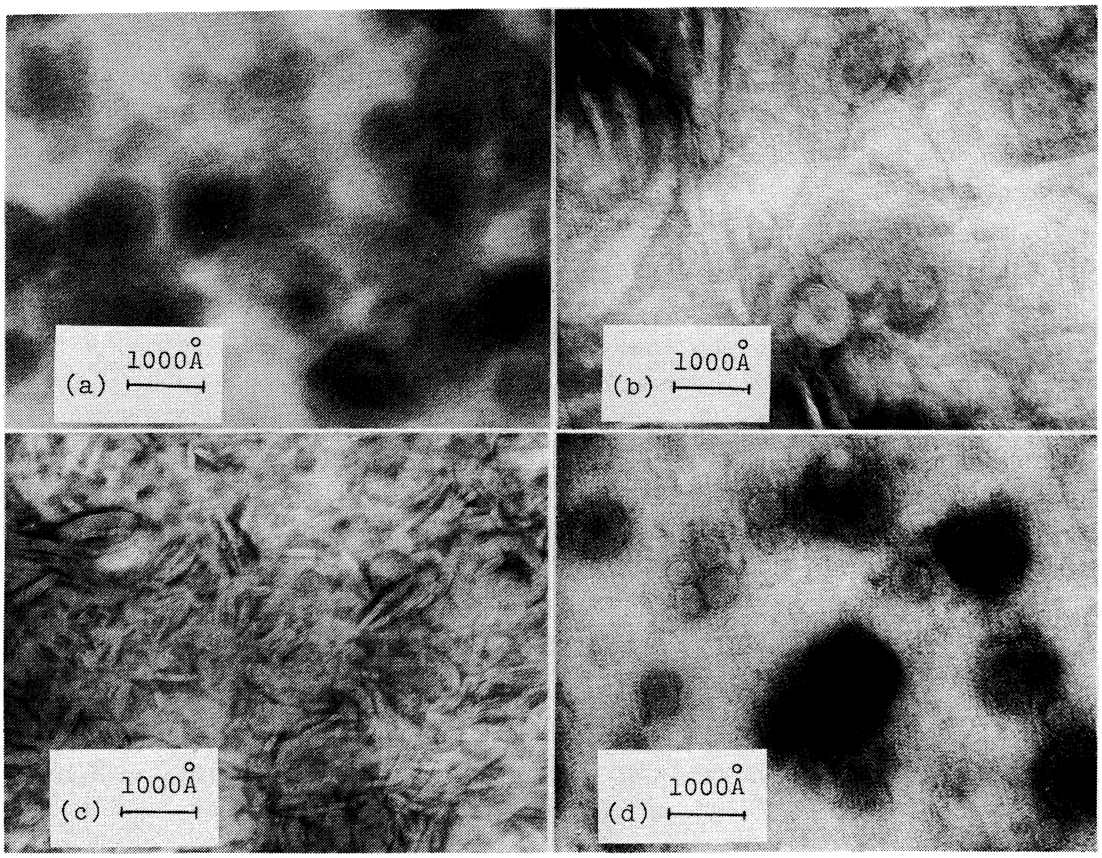

Fig. 1 Electron micrographs of PVA film containing artificial lipids
a) 1 / PVA = 60 wt%, b) 2 / PVA = 80 wt%, c) 3 / PVA = 100 wt%,
d) 1 / PVA = 60 wt%, 4 / 1 = 50 mole%

References

1) T. Kunitake, Y. Okahata, K. Tamaki, F. Kumamaru, and M. Takayanagi, Chem. Lett., 1977, 387.
2) T. Kunitake and Y. Okahata, Chem. Lett., 1977, 1337.
3) T. Kunitake and Y. Okahata, Bull. Chem. Soc. Jpn., 51, 1877 (1978).
4) T. Kajiyama, A. Kumano, M. Takayanagi, Y. Okahata, and T. Kunitake, Chem. Lett., 1979, 645.
5) T. Kunitake and S. Yamada, Polym. Bull., 1, 35 (1978).

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