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Note

Effect of sucrose on the structure of a cubic phase formed from a monoolein/water mixture

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Abstract

The effect of sucrose on the structure of molecular assemblies formed in an MO/H_2O mixture has been studied using a small-angle X-ray scattering method. It was found that the phase transition *Ia3d* → *Pn3m* → H_{II} occurs with increased sucrose concentration in the mixture with the composition 70 wt% MO at 20 °C. This structural change induced by sucrose addition would be ascribed to a cosmotropic property of sucrose, which leads to the dehydration of the MO head group and hence causes a decrease in the effective area occupied by the MO molecule at the polar/apolar interface, which facilitates the formation of molecular assemblies with a high curvature for the reversed liquid-crystal mesophase.

Keywords: Cubic phase; Hexagonal phase; Monoolein; Phase transition; SAXS

1. Introduction

Lipids are important materials in view of their wide applications in many fields such as pharmaceuticals, food processing, the paper and pulp industry, and the dairy and fine chemical industries. In pharmaceutical applications mixture with additives, such as free fatty acids and carbohydrates, as well as with added surfactant molecules will alter the structure of the lipid aggregates and the characteristics of the lipid–aqueous interface.

The phase behavior of monoolein (MO)/H_2O mixtures has been studied intensively [1–7]. It is well established that at 20 °C, two reversed bicontinuous cubic phases are formed depending on the water content in the mixture [4,7,8]. One is the cubic phase with space group type *Ia3d* formed in the mixture with 19–38 wt% water, and the other is with space group type *Pn3m* formed in the mixture with 37–42 wt% of water. At higher water content, the latter cubic phase still coexists with excess water. When the temperature is increased to 80 °C, the reversed hexagonal phase (H_{II}) appears, which can also coexist with excess water [9,10].

On the basis of investigations performed to date [1,4,11–14], the reversed cubic phases of the MO/H_2O mixture are considered to be built up of a three-dimensional network of curved lipid bilayers separated by a network of congruent water channels. For these cubic mesophases, the center of the lipid bilayer is expected to lie on an infinite periodic minimal surface (IPMS) of hyperbolic shape. Specifically, the *Ia3d* and *Pn3m* cubic phases can be represented by the "gyroid (G)"- and "diamond (D)"-type IPMS, respectively.

The cubic phase has a transparent, stiff, and gel-like appearance that often limits its potential use in drug delivery systems. It would be convenient for practical applications if the phase behavior of the MO/H_2O mixture could be modified by additives to exhibit a hexagonal phase, because the less viscous hexagonal phase must result in several opportunities for routes of administration and applications as a drug delivery system. Furthermore, coexistence with excess water is characteristic of a reversed hexagonal phase as well as

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of a reversed cubic phase, which is an advantage of these mesophases for wide use in practical applications such as in the food industry and in drug delivery systems [15,16]. In the present study, we investigated the effect of sucrose on the structure of molecular assemblies formed in an MO/water mixture with a composition of 70 wt% MO at constant temperature 20 °C. The purpose of this work is to explore the possibility of modifying the MO/H_2O phase behavior to obtain more convenient mesophases for practical application by the use of additives.

2. Experimental

2.1. Materials and sample preparation

Monoolein (1-monooleoyl glycerol, MO, purity >95%) was purchased from Danisco Cultor (Denmark). Sucrose (purity >99%) was purchased from Sigma–Aldrich. These materials were used directly without further purification. All the samples were prepared using double-distilled water.

Liquid-crystal samples containing MO, water, and sucrose were prepared as follows. In the typical case with mass ratio MO:H_2O = 7:3, sucrose was added in various weight ratios (less than 30%) to the MO/H_2O mixture. All the samples were stored at 50 °C for 24 h until the mixture came into a homogeneous phase; then they were kept at room temperature for 40 days in order to reach equilibrium.

2.2. Small-angle X-ray scattering

SAXS measurements were performed using a Kratky compact camera (HMBG, Austria) with Ni- and W-filtered Cu$K\alpha$ radiation (wavelength $\lambda = 0.154$ nm) generated by a PW3830 X-ray generator (5 kV × 40 mA). Scattering intensities were plotted against reciprocal spacing $S = 2\sin\theta/\lambda$, where 2θ is the scattering angle. Exposure time was taken to be 1000 s for all samples.

3. Results and discussion

At 20 °C and with 30 wt% water content, MO forms a reversed cubic phase with space group *Ia3d* (Q230) [17]. The effects of sucrose on the structure of the MO/water cubic phase were investigated by the SAXS method at 20 °C as a function of sucrose concentration.

In the SAXS pattern of the MO/water/sucrose mixture containing 1.0 wt% sucrose, 28.2 wt% H_2O, and 70.8 wt% MO (Fig. 1a), there appear two main peaks, the positions of which correspond to the S values of 0.233 nm^{-1} (S_1) and 0.269 nm^{-1} (S_2). The ratio of these two peaks is $\sqrt{6}$:$\sqrt{8}$, which suggests that the two diffraction peaks correspond to (211) and (220) reflections on a three-dimensional cubic phase of a space group *Ia3d* [17]. The reciprocal spacing, S, of the cubic phase is connected with the lattice constant, a_0, by $S_{(h,k,l)} = (1/a_0)(h^2 + k^2 + l^2)^{1/2}$, where h, k, and l are Miller indices [4]. For the correct choice of space group, this plot gives a straight line passing through the origin and having a slope equal to the reciprocal unit cell axis ($1/a_0$) (see Fig. 2).

Addition of more sucrose into the MO/H_2O system changes this cubic structure. As shown in Fig. 1b, the SAXS pattern of the 5.0 wt% sucrose/66.8 wt% MO/28.2 wt% water mixture exhibits four main peaks, the positions of which correspond to the S values 0.191 nm^{-1} (S_1), 0.220 nm^{-1}

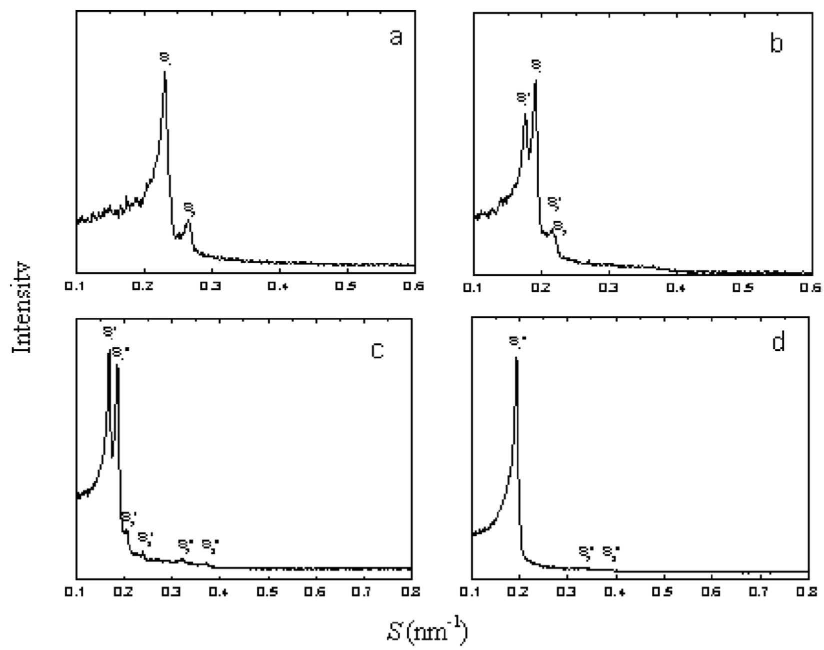

Fig. 1. X-ray scattering profiles of MO/H_2O/sucrose mixtures with mass ratio MO:H_2O = 7:3 at 20 °C. 1.0 wt% sucrose (a), 5.0 wt% sucrose (b), 10.0 wt% sucrose (c), 15.0 wt% sucrose (d).