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Molecular Arrangement of Lamellar Liquid Crystalline Structure with Virgin Coconut Oil

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The molecular arrangements of lamellar mesophase of a mixed surfactant system with decanol were studied, before and after the addition of virgin coconut oil (VCO) by employing optical microscope and small-angle X-ray scattering (SAXS). The two methods have enjoyed much success in elucidating information on the molecular arrangements of liquid crystals. The mixed surfactant used are dodecyl trimethylammonium bromide (DTAB) and aerosol-OT (AOT) at a weight ratio of 90:10. The results from the optical pattern show that the water/DTAB:AOT (90:10)/decanol systems were able to solubilized only small amount of virgin coconut oil before transforming into other structures. From the small-angle X-ray scattering spectrum, it was found that the virgin coconut oil molecules to be partitioned between the nonpolar methyl group layers of lamellar liquid crystalline structure. This was indicated by the higher *d*-spacings value upon addition of virgin coconut oil. Finally, the water molecules located between the polar head groups, showed low penetrating tendency into the layered structure of the mixed surfactants system.

Keywords: Virgin coconut oil, Surfactants, Lamellar liquid crystal, Small-angle X-ray scattering.

INTRODUCTION

Ordinary coconut oil was not much sought after oil by the industry due to the limited options available. It was not popular due to its oil content containing moisture, high saturated and trans fats. However, of late with the introduction virgin coconut oil (VCO), it is considered as the gifted crop and its choices are abundant. Today, virgin coconut oil is the oil for beauty and health [1]. Its application are vast as in pharmaceuticals, nutriceuticals and cosmeceuticals and health foods. It is a clear oil with distinct aroma and unique flavour when it is in its virgin form [2]. It is a vegetable oil obtained from the kernels of the coconut palm also known as *Cocos nucifera*. The difference between an ordinary coconut oil and virgin coconut oil lies in the process of extraction. Ordinary coconut oil can be extracted either through dry or wet processing with a certain amount of water content. While virgin coconut oil is obtained from the coconut milk of fresh coconuts through wet processing and is more desirable because there is no chemical or heating is imposed on the oil. Thus virgin coconut oil is free from moisture and is more environmental friendly [3]. Virgin coconut oil is known for its medium chain triglycerides (MCT) and high content of lauric acid (about 50 %). *In vitro* studies showed that these medium chain fatty acids and its corresponding monoglycerides have antifungal, antibacterial, antiviral properties including human pathogens like herpes simplex virus and antiprotozoal [4-8].

Lamellar liquid crystal as the name implies is a layered mesophase structure that flows like a liquid but solid like a crystal. It is an interesting colloidal association structure that can be observed in many phase diagram consisting of water/co-surfactant/surfactant systems. It has been reported that presence of this structure is the main reason behind the stability of many products such as in the cosmeceuticals skin care industries. Hence its structure together with the location and molecular arrangement of the compounds in the structure becomes crucial and tantamount in understanding its behaviour and properties [9-12]. In this work we present the optical pattern and small-angle X-ray scattering measurements to indicate the perturbation of the lamellar structure upon addition of virgin coconut oil as well as the partition and location of virgin coconut oil molecules in the different part of the lamellar structure. This information is useful for many formulation work and especially in many industrial applications [13].

EXPERIMENTAL

The chemicals were purchased from Sigma Aldrich and Merck (> 99 %) while the virgin coconut oil was purchased from the Wellness Connection Sdn. Bhd., Malaysia. All of the

chemicals were used directly without undergoing further purification. Meanwhile, the water used throughout the studies underwent double distillation. The composition of virgin coconut oil and its properties were reported elsewhere [13,14].

Construction of phase diagrams: The basic idea is to understand and predict what phases will be stable in a system when the compositions are defined and how those stability relations will change as these compositions are varied. The phase diagrams were determined on a clear/turbid criteria basis by mixing two of the components and titrating with the smallest amount of the third component. The samples were then thoroughly mixed to homogeneity with a vortex mixer, centrifuged and then allowed to reach equilibrium at a specific temperature in a water bath. The phases were then examined by visual inspection between cross polarizers. An estimated region of the phases can then be made by this method by noting the turbid and clear compositions.

Polarized optical microscope: The liquid crystalline phases were studied and differentiated by polarizing optical microscopy. The images were captured by Olympus Provis AX70 equipped with polarizing filters and PM30 automatic photomicrography. A small amount were transferred on microscope slide and sheered immediately with glass. Sample slide was placed under the stage. Magnification of 200x were used and photomicrographs of various optical pattern were captured by the camera.

Small angle X-ray scattering: The small angle X-ray scattering measurements were performed on the Hecus-Braum small and wide angle X-ray scattering facility at the Nuclear Science Building in National University Malaysia (UKM). Nickel filtered CuK_α radiation generated from a cooper anode operating of 40 kV and 20 mA was used as the radiation source by PANalytical PW3830 X-ray Generator. Samples were loaded into a stainless steel holder whereby the sample to detector distance was 274 nm. The measurements were carried out at room temperature (25 °C) and at the wavelength of 0.154 nm with measuring time of 1000 s. The interlayer spacing of the liquid crystalline was obtained from the first scattering peak and this value was used to determine the microstructure of the lamellar liquid crystalline. Virgin coconut oil at 3 and 5 weight percent were then added to the lamellar samples and followed similar procedure to measure the interlayer spacing.

RESULTS AND DISCUSSION

Phase diagram water/decanol/AOT:DTAB (10:90): Results showed the existence of two region of homogenous isotropic dispersion as shown in Fig. 1. One in the part of high aqueous content and the other with high alcohol content. These regions are the micelle and inverse micelle region, respectively. The limits of these area were determined by titrating to turbidity with the smallest amount of one component to the homogenous dispersion. The formation of lamellar liquid crystalline region was detected when 20 % water was added onto the surfactants mixture and the region expanded progressively with the addition of water (Fig. 1). At the alcohol-free axis, the bilayer structure were able to solubilize approximately 45 % water by weight. This region were also able to solubilize as much as 17 % decanol with a maximum water solubility of 64 % by weight. This

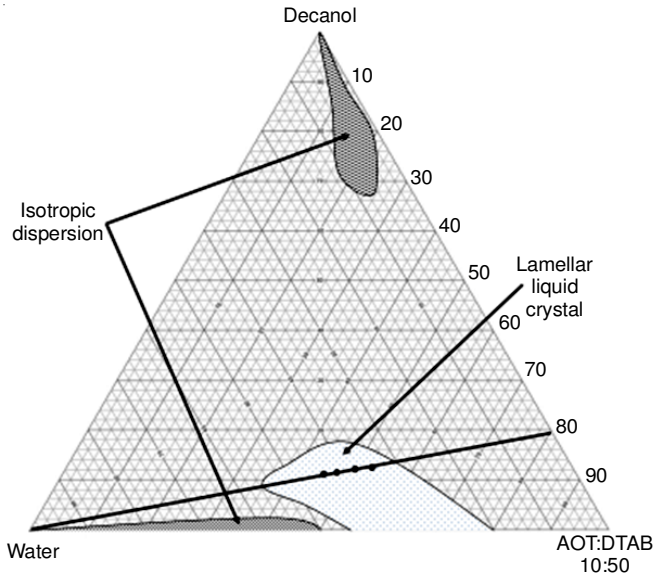

Fig. 1. Pseudoternary phase diagram of water, decanol and mixture of aerosol-OT and DTAB at weight ratio of 10:90

region was further verified under polarized microscope to observe their optical pattern.

Optical pattern: The optical pattern of the liquid crystal was taken at the dotted points of Fig. 1, corresponding to 35-45 by weight per cent of water. The systems consisting of mixture of surfactants/decanol exhibited a typical lamellar liquid crystalline structure with water contents in the range investigated. As a way of illustration the optical pattern at these compositions when observed under a polarizing microscope are given in Fig. 2a-d, with and without virgin coconut oil at variable water content. Addition of the mixed surfactant and with the alcohol gives a typical lamellar with oily streaks at the water fractions investigated (Fig. 2a-b). However, when virgin coconut oil was added to the lamellar structure, the optical pattern takes the pattern of oily streaks with slight maltese crosses present (Fig. 2c-d).

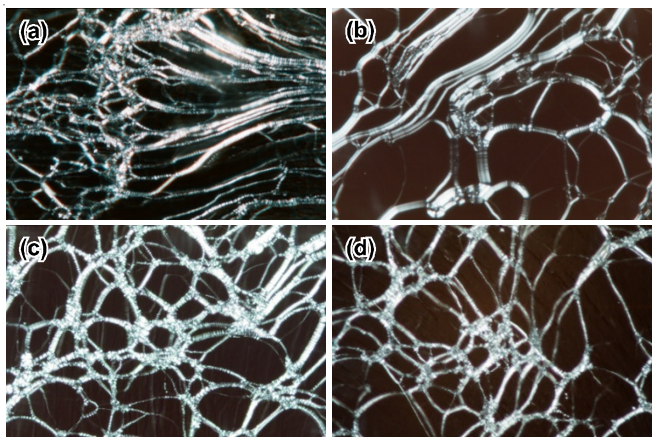

Fig. 2. Optical patterns of systems (a,b) without and with (c,d) virgin coconut oil at variable water content

Small angle X-ray scattering: The effect of the solubilization of virgin coconut oil on the dimension of lamellar liquid crystalline structure was determined by small angle X-ray scattering (SAXS). The measurements were made for each

sample with the composition as shown by the dotted pointed in Fig. 1 followed by similar sample with addition of virgin coconut oil. The typical small angle X-ray scattering spectra are shown in Fig. 3 for the lamellar liquid crystal samples without virgin coconut oil at variable water content.

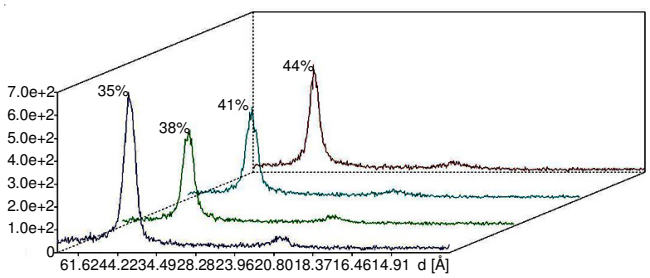

Fig. 3. Small angle X-ray scattering spectrum of lamellar mesophase of dotted points in Fig. 1

The interlayer spacings of each lamellar liquid crystalline sample were then calculated from the respective small angle X-ray scattering spectrum. The results are then illustrated as shown in Fig. 4, before and after addition of virgin coconut oil plotted against water ratio in the range of 35-45 by weight per cent, corresponding to the weight ratios of 0.5-0.8. The interlayer spacing of the lamellar system without virgin coconut oil increased from 40.9 to 43.9 Å in the interval used (dotted points of Fig. 4).

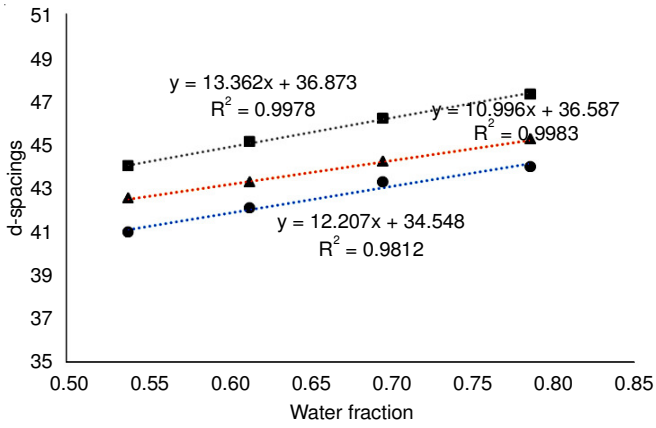

Fig. 4. Interlayer spacing as a function of water fraction before, ● addition of virgin coconut oil and after addition of 3, ▲ and 5 wt %, ■, virgin coconut oil, respectively

Upon addition of 3 weight per cent of virgin coconut oil (solid triangle of Fig. 4), increased the interlayer spacing slightly by about 2 Å as well as slight increase in the value of the slope. Subsequent addition of 5 weight per cent virgin coconut oil (solid square of Fig. 4), a similar pattern was observed, *i.e.*, it showed increasing the water content, the lattice spacing of the lamellar liquid crystalline also increased. It suggested that the presence of virgin coconut oil in the lamellar liquid crystalline affected the lattice spacing where the interlayer spacing increased as the virgin coconut oil content increased.

From the figure, it was observed that the curves for system with and without virgin coconut oil were almost parallel to each other with approximately 2 Å apart but with slight variation on the slopes. The interlayer spacing observed from the small angle X-ray scattering patterns provides additional information about the organization in the lamellar liquid crystalline structure after calculation of the interlayer spacings in the amphiphilic layer (palisade) and water layer separately. The thickness of the amphiphilic layer can be obtained by extrapolation of the interlayer spacing in Fig. 4 to zero water content. With that information available, the partitioning of water may be calculated. Table-1 summarized the extrapolated values obtained from the plots. The data in Table-1 revealed that there was a regular increased in the extrapolated values of interlayer spacing (d_0) for all systems with and without virgin coconut oil.

TABLE-1
EXTRAPOLATED INTERLAYER SPACING VALUES, d_o

Surfactant system	d_0 values (Å)		
	0 % Virgin coconut oil	3 % Virgin coconut oil	5 % Virgin coconut oil
DTAB+AOT (90:10)/C10-OH/H_2O	34.55	36.59	36.87

The partitioning of water can be characterized by the penetration fraction, α and is defined by Hamdan *et al.* [14]:

$$d = d_o [1 + (1 - \alpha)R]$$

The calculated values are tabulated as shown in Table-2. The comparison between the d_o values, which are the measure of the amphiphilic part of the structure, before and after the addition of virgin coconut oil were informative when compared to the calculated d_o value of fully extended DTAB molecules of 16.7 Å [15]. The extrapolated spacing was found to be in excess of the value calculated fully extended chains. This can be explained by i) due to the presence of bulky double hydrocarbon chain in the aerosol-OT molecule in the amphiphilic layer. The stearic hindrance exerted by the anionic charged and bulky aerosol-OT may have pushed the DTAB molecule upward and inducing a temporary disorder of lamellar structure. Hence, extending the amphiphilic layer thickness and ii) the amphiphilic chains being located in the space between the methyl group layers.

TABLE-2
PENETRATION FRACTION (α) OF WATER

Surfactant system	Penetration fraction (α)		
	0 % Virgin coconut oil	3 % Virgin coconut oil	5 % Virgin coconut oil
DTAB+AOT (90:10)/C10-OH/H_2O	0.622	0.611	0.644

Further increased in the d_o values after the addition of virgin coconut oil suggested that the virgin coconut oil molecules to be located between the methyl group layers which is the nonpolar region of the structure. The calculated penetration fraction of the water (Table-2) showed that there was little penetration of water into the amphiphilic layers which is satisfying since water is expected to be located at the polar region of the structure.

Conclusion

The results gave evidence that the dimensions of the lamellar liquid crystalline were affected by the presence of

virgin coconut oil as indicated by the dislocation in the optical pattern and the variation in the interlayer spacings of the lamellar structure. The interlayer spacing obtained from the small angle X-ray scattering spectrum was able to provide information on the molecular organization with the liquid crystalline. From the interlayer spacing values extrapolated to zero water content revealed the thickness of the amphphilic layer. From the results it can be deduced that the virgin coconut oil molecules were localized in between the methyl (non-polar) layer with certain degrees of penetration into the lipophilic chain in the amphphilic layers. In addition, the presence of virgin coconut oil into the system further increased the thickness of the amphiphilic layer. However, the presence of virgin coconut oil hinders penetration of water.

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