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by Ananda Putra

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Solubility dynamic of methyl yellow and carbon black in microemulsions and lamellar liquid crystal of water, non ionic surfactants and cyclohexane system

A Amran*, R Harfianto, W Y Dewi, D Beri and A Putra
Department of Chemistry, Faculty of Mathematics and Sciences, Universitas
Negeri Padang, Jl. Prof. Dr. Hamka, Air Tawar Barat, Padang, Sumatera
Barat, Indonesia 25131

*E-mail: amrana.unp@gmail.com

Abstract. Solubility dynamics of methyl yellow and carbon black in microemulsions and liquid crystals of water, non-ionic surfactants and cyclohexane system, have been investigated. Actually, solubility dynamics of these dyes both in microemulsion (w/o microemulsions) and the lamellar liquid crystal (LLC) were strongly related to the chemical composition, nature and characteristics of microemulsions and the lamellar liquid crystals.

1. Introduction

Interaction among organic dyes and surfactants became an interested and technological research areas [1-4], since organic dyes can be able to probe of association colloid [5-6]. Solubility area of organic dyes has been investigated and focus on micellar solution, whereas solubility area has been limited studied [7, 8]. Additionally, it has been evaluated the hydrophilic-liphophilic equilibrium. The goal of this research at getting the interface interaction that can be able to compare more specific interface study [9, 10]. In fact, this fundamental knowledge will be useful and will be able to understand the thermodynamic and general solubility rules [11]. This information is also important to explain and establish the relationship between colloidal structures in microemulsion system and the organic dyes solubilities [12, 13]

Solubility of organic pigments in microemulsions and liquid crystals systems have importantly scientific and technological values which can be able to apply spontaneously in industries of paints [14], inks and pharmacies [15], cosmetics and some other manufactures [16]. Information about physical and chemical properties can be got from mapping of diagram phases in some needed physical and chemical systems [17, 18]. By doing so, it can be able to make improvisation or to more increased application values of non-ionic surfactants.

In fact, dynamics solubility in water, non-ionic surfactants, and cyclohexane system became more interesting area, because the system, at least, performing four surfactant association structures i.e. normal micel, and inversed micel (oil in water, o/w and water in oil, w/o microemulsions, in general term), lamellar liquid crystal, and hexagonal liquid crystal. The four surfactant association structures have different physical and chemical properties, as well [17].

This study aimed at knowing solubility dynamics of methyl yellow and carbon black in microemulsions and lamellar liquid crystal regions. Methyl yellow was able to get: "a permanently red colour in the water of pH = 1, and a permanently yellow colour in the water pH =



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5. Whereas, fine divided carbon black was able to get a black colour. Ultimately, permanently red, yellow, and black colours of microemulsions and LLC will be used to fulfil ballpoint ink and cartridge printer.

2. Experimental section

2.1. Materials and Method

The following chemicals were used without further purification. Non-ionic surfactants (Tween-20, Tween 40, Tween 60, and Tween 80) and cyclohexane, were purchased from Merck, Germany. Methyl yellow and fine divided carbon black were purchased from Wako Pure Chemical Industries Ltd. Double distilled water was purchased from Rafi Medika. Nitric acid (70%, ACS certified, Baxter Healthcare Corp, Scientific Product Division) was used as acid medium to adjust the water to a pH = 1, and pH = 5.

2.2. Phase diagram preparation

The phase regions of microemulsions and liquid crystals were obtained by titration of cyclohexane/non-ionic surfactant with the water (pH = 1, and pH = 5). The phase region limits were determined by visual observation between crossed polarizers with parafilms and was confirmed by the results from Hund-Wetzlar® optical polarizing microscope.

2.2.1. Solubility of Methyl Yellow and Carbon Black in Microemulsions and Lamellar Liquid Crystals. Solubility of methyl yellow in w/o microemulsions and lamellar liquid crystals was determined for system of water (pH = 1, and pH = 5), Tweens (Tween-20, Tween-40, Tween-60, and Tween-80), and cyclohexane system. Solubility of methyl yellow and fine divided carbon black in hexagonal liquid crystal was not determined. The procedure used: a small quantity of methyl yellow and fine divided carbon black were added gradually into a tube which already filled with given compositions of microemulsions and/or lamellar liquid crystals phases of interested samples. Then, the mixture of samples was mixed using Vertexer. Hence, the homogeneity of dyes solubility was observed visually using Red laser light and the Hund Weslar. The addition of dyes was stopped when a precipitation of dyes appeared.

3. Results and Discussion

Figure 1 showed the structure of Tween 40 as an example of Tween compounds that can be widely used as emulsifier, solubilizers, and wetting agents. The solubility of Tween aqueous solutions increased with the degree of ethoxylation. For a fixed degree of ethoxylation, aqueous solubility decreased as the number of ester groupings increased. For fixed degree of ethoxylation and esterification, aqueous solubility decreased as the molecular weight of the fatty acid increased.

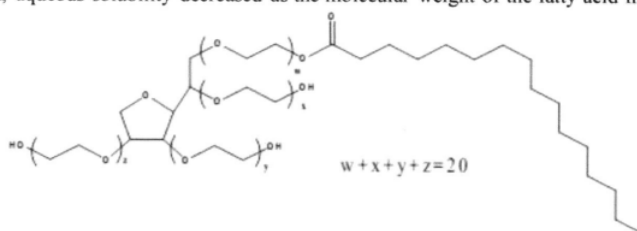


Figure 1. Example of Tween structure (Tween-40)

In this research, methyl yellow and carbon black were used as dyes. Methyl yellow was a chemical compound which might be used as pH indicator. In aqueous solution at low pH, methyl yellow

appeared red. Between $\text{pH} = 2.9$ and $\text{pH} = 4.0$ methyl yellow underwent a transition, to became yellow above $\text{pH} = 4$. So, methyl yellow was able to get red colour in water of $\text{pH} = 1$, and yellow colour in water of $\text{pH} = 5$. Figure 2 showed the structure of methyl yellow. Furthermore, carbon black was able to get a black colour in water ($\text{pH} = 1$ and $\text{pH} = 5$).

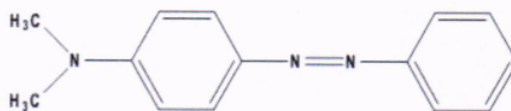


Figure 2. Structure of methyl yellow

3.1. Phase diagram

The results were interesting phase diagrams of water ($\text{pH} = 1$), non-ionic surfactants (Tween-20, Tween-40, Tween-60, and Tween-80), and cyclohexane system, was performed in figure 3. It was shown from this figure that the performance of surfactant association structures, i.e w/o microemulsions, lamellar liquid crystals (LLC) areas formed larger by increasing the total number of moles of ethylene oxide (20 to 80) of Tweens. Except for Tween-60 and Tween-80, the surfactant association structures of hexagonal liquid crystals were also formed.

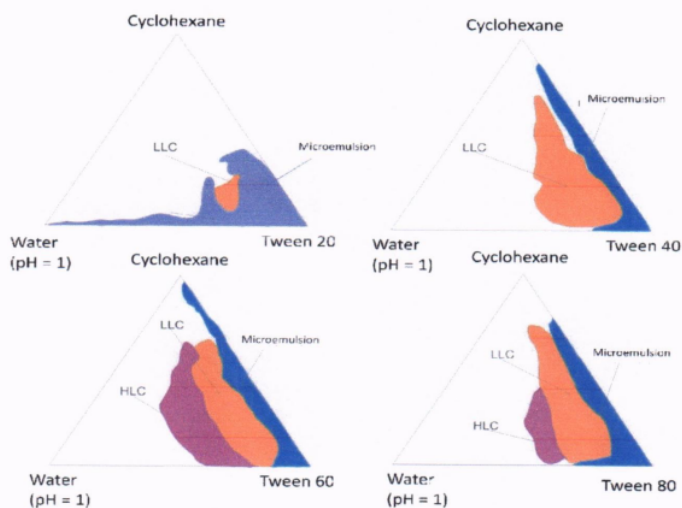


Figure 3. Phase diagram of water ($\text{pH}=1$), Tweens (Tween-20, Tween-40, Tween-60, and Tween-80), and Cyclohexane System.

The phase diagrams of water ($\text{pH} = 5$), non-ionic surfactants (Tween-20, Tween-40, Tween-60, and Tween-80), and cyclohexane system, was performed in figure 4 which relatively similar to the phase diagrams of water ($\text{pH} = 1$) non-ionic surfactant (Tween-20, Tween-40, Tween-60, and Tween-80), and cyclohexane system. It meant that the exchanging of pH (from 1 to 5) of water would not exchange the formation of the surfactant association structures.

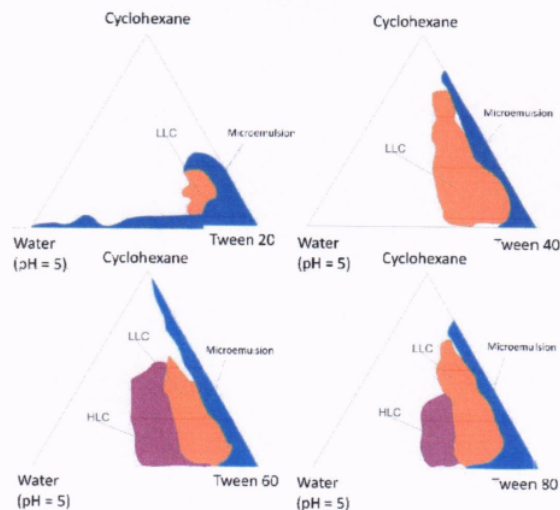


Figure 4. Phase diagram of water (pH=5), Tween (Tween-20, Tween-40, Tween-60, and Tween-80), and Cyclohexane System.

3.2. Solubility of methyl yellow and carbon black in microemulsion and lamellar liquid crystals

Table 1 showed the solubilities of methyl yellow and carbon black in the water (pH = 1, and pH = 5), non ionic surfactant (Tweens) and cyclohexane system. The solubilities of both methyl yellow and carbon black were not exchanged by changing the pH of water from 1 to 5 in the region of w/o microemulsions. It just maintained colour stability of methyl yellow and carbon black. The solubilities of methyl yellow in the w/o microemulsion regions which involved Tween 20 was higher than the other Tweens, because in that region the surfactant contents were higher.

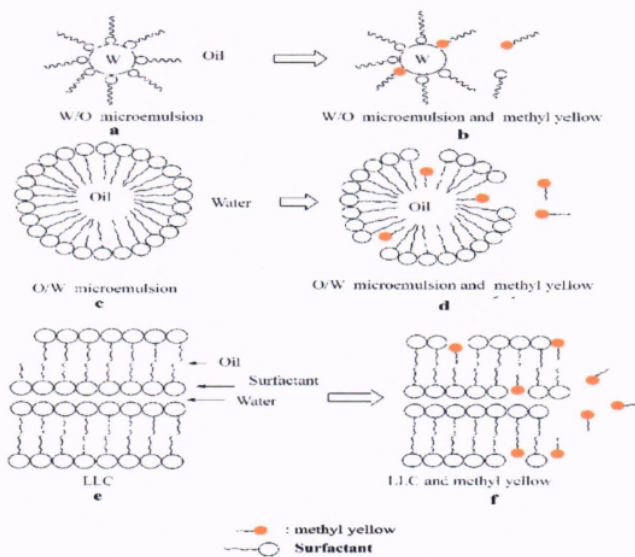
On the other hand, the solubilities of methyl yellow in the region of LLC were lower than in that of w/o microemulsion regions, because the physical structure of LLC was more compact than physical structure of w/o microemulsion as shown in the figure 5. In the system with the water of pH=1, the solubilities of methyl yellow were lower than that of the system with the pH = 5. These might water of be due to the physical structure of LLC became less compact.

The solubilities of carbon black in both association structures' regions, w/o microemulsion and LCC, and both water pH (pH = 1 and pH = 5), were too low (less than 0.2 mg/g). It indicated that the solvent medium was not suitable.

Table 1. The solubilities of methyl yellow and carbon black in the water (pH = 1, and pH = 5), non ionic surfactant (Tweens) and cyclohexane system

Surfactants	Regions	Dyes (pH=1)		Dyes (pH=5)	
		Methyl yellow (mg/g)	Carbon black (mg/g)	Methyl yellow (mg/g)	Carbon black (mg/g)
Tween-20	w/o microemulsion	1.6	<0.2	1.6	<0.2
	LLC	<0.2	<0.2	1.2	<0.2
Tween-40	w/o microemulsion	0.4	<0.2	0.466	<0.2
	LLC	<0.2	<0.2	0.284	<0.2
Tween-60	w/o microemulsion	<0.2	<0.2	<0.2	<0.2
	LLC	0.4	<0.2	0.6	<0.2
Tween-80	w/o microemulsion	0.266	<0.2	0.666	<0.2
	LLC	0.17	<0.2	0.638	<0.2

The solubility of methyl yellow in w/o microemulsion, o/w microemulsion, and lamellar liquid crystal, was proposed as shown in Figure 5. It was seen that the polar head of methyl yellow would dissolved in polar medium, like water. The non-polar ring (as a tail) of methyl yellow, on the other hand, would dissolved in oil phase.

**Figure 5.** Solubility illustration of methyl yellow

4. Conclusions

It concluded that solubility dynamics of methyl yellow and carbon black, both in w/o microemulsions and the lamellar liquid crystal (LLC), were strongly related to the chemical composition, nature and characteristics of microemulsions and the lamellar liquid crystals. In the system with the water of pH = 1, the solubilities of methyl yellow were lower than that of the system with the water of pH = 5. These might be due to the physical structure of LLC became less compact. Whereas, the solubilities of carbon black in both association structures' regions, w/o microemulsion and LCC, and both water pH (pH = 1 and pH = 5), were too low. It indicated that the solvent medium was not suitable, yet.

Acknowledgement

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