PHASE BEHAVIOR STUDY OF VEGETABLES OILS WITH TWEEN85 AND TWEEN80 IN EMULSION SYSTEM

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Abstract - In this research, new emulsions were prepared based on three different vegetable oils which are sunflower oil, virgin coconut oil and extra virgin olive oil with different non-ionic surfactant, namely Polyoxyethylene (20) sorbitanmonooelate (Tween 80) and Polyoxyethylene (20) sorbitan tri-oleate (Tween 85). Phase behaviors of vegetables oil/non-ionic surfactants/water were investigated. The effects of the surfactants on the emulsion system of the phase were elucidated. Thus, ternary phase diagram were constructed to find the suitable and stable emulsion system of vegetable oils/non-ionic surfactant/water for cosmeceutical purposes.

I. INTRODUCTION

Nowadays, products derived from vegetable oils, which are considered important substances obtained from renewable resources. Vegetable oils are tri-esters of straight-chained mostly unsaturated fatty acids with glycerol, with higher levels of biodegradability and lower toxicity than conventional mineral or synthetic oils. Sunflower oil is the non-volatile oil compressed from sunflower seeds where it is commonly used in food as frying oil and in cosmetic formulations. It has higher amount of unsaturated fat and lower amount of saturated fat which contain of vitamin E, sterols, squalene, and other aliphatic hydrocarbons, terpene and methyl ketones. Besides that, Virgin Coconut Oil (VCO) is also one of the major natural products used in cosmeceutical products. It is effective base oil for composition within the lotion, cream, body butter and lip products formulations. While olive oil contains 56-87% of monounsaturated fatty acid, 8-25% of saturated fatty acids and 8-22% of polyunsaturated fatty acids (Koutsopoulos et al., 2008). It has a unique colour due to its pigments like chlorophyll, pheophytin, and carotenoids. A surfactant is a compound that lowers the surface tension of a liquid, increasing the contact between the oil and water. They are the compounds that an interface could reduce the interfacial tension (Miller et al., 2001), and have been extensively used in consumer products such as cosmetics and foods. Surfactants facilitate the preparation of an emulsified system by lowering the free energy in the system and by reducing the work required to generate the new interfaces (Myers, 2006). The surfactant molecule lowers the interfacial tension between the oil and water phases. In this research, phase behavior of vegetable oils with surfactants was investigated in order to obtain stable emulsion.

II. MATERIALS AND METHODS.

2.1 Materials
Sunflower Oil was obtained from Lam Soon Edible Oils Sdn. Bhd., Malaysia. It contains of 884 kCal of energy, 11% saturated fat, 20% monounsaturated fat and 69% polyunsaturated fat for every 100mL of serving. Virgin Coconut Oil (VCO) was purchased from Wellness Connections Sdn. Bhd., Malaysia. It contains of 675.4 kCal of energy, 4.2% carbohydrates, 0.6% proteins and 95.2% fat for every 100mL of serving. Extra Virgin Olive Oil was obtained from Bertolli, Italy. For every 100mL of Extra Virgin Olive Oil serving, the amount of energy is 856.8 kCal, saturated fat is 9%, monounsaturated fat is 10% and polyunsaturated fat is 2%. Polyoxyethylene (20) sorbitan tri-oleate (Tween 85) and Polyoxyethylene(20) sorbitan mono-oleate(Tween 80) were purchased from FlukaChemie GmbH, USA.

2.2 Construction of Ternary Phase Diagram

2.2.1 Phase Behavior of Vegetable Oils
Vegetable oil/Nonionic Surfactant/Deionized Water were weighed in the range of 0:100 to 100:0 (Vegetable Oil/Nonionic Surfactant/Water (w/w)). 0.5g of mixture was weighed and then placed in a 10mL screw-cap glass tube. 11 test tubes were required for each type of oil and surfactant. The samples were mixed by using vortex mixture for 5 minutes and being centrifuged for 15 minutes at 4000 rpm. The phase behaviors of the samples were examined through cross-polarized light. The experiment was repeated with the addition of deionized water according to its percentage from 0%
to 100%. The phase behaviors were determined from visual observation as shown in figure 3.3. Ternary phase diagram was drawn accordingly.

2.2.2 Effect of Phase Behavior of Vegetable Oil with Respect to Different Nonionic Surfactants

Two nonionic surfactants were selected to construct ternary phase diagram of vegetable oils. They were Polyoxyethylene(20) sorbitan tri-oleate (Twee 85) and Polyoxyethylene(20) sorbitan mono-oleate (Twee 80). The Hydrophilic-Lipophilic Balance (HLB) values of Twee 85 and Twee 80 are 11.0 and 15.0 respectively. Comparison among the ternary phase diagrams were made to see the changes that happened when the different HLB values of the nonionic surfactant were used.

III. RESULTS AND DISCUSSION

3.1 Phase Behavior Study of Vegetable Oils with Respect to Different Non-Ionic Surfactants

The mixture is containing of oil, water and surfactant in different ratio able to produce different types of emulsion systems. They depend on the chemical nature, molecular structure and concentration of surfactants and other ingredients (Lakatos-Szabo et al., 2007). Non-ionic surfactants such as Twee 80 and Twee 85 series are stable surfactants and safe agents for all biological tissue in general and for skin in specific (Whitehead K et al., 2007). These non-ionic emulsifiers are compatible with various ingredients used in the preparation of emulsions and are not affected by pH. They supposed to have an enhancement effect on the skin barrier (Fang et al, 2001). Construction of ternary phase diagrams is the best way to study all types of formulation.

Six ternary phase diagrams were constructed in the study of phase behavior of vegetable oils with respect to different non-ionic surfactants. Each of the phase diagrams is representing the phase diagram of water, vegetable oil and surfactant with different HLB values. The vegetable oils were Sunflower Oil, VCO and Olive Oil. Then, the surfactants were Twee 85 and Twee 80 having HLB values of 11.0 and 15.0. The changes of phase behavior were observed in the ternary phase diagram.

3.2 Phase Behavior of Twee 85/Sunflower Oil/Deionized Water

Figure 3.2 shows the ternary phase diagram of Twee 85/Sunflower Oil/Deionized Water. The regions that appeared in this ternary phase diagram were two phase (T) and homogeneous (H) regions. The homogeneous region, milky emulsion system was formed along the apex line of Twee 85 and Sunflower Oil. However, increasing the percentage of deionized water caused the formation of two-phase region. The two-phase region was found at the water-rich corner in the Twee 85/Sunflower Oil/Deionized Water system. The two phase regions also appeared when using Twee 80 as an emulsifier. There is no isotropic region found in Twee 85/Sunflower Oil/Deionized Water system. In Twee 85, two hydroxyl groups of the polyethylene moiety of Twee 80 are substituted by two lipophilic oleate tail groups and form polyoxyethylenesorbitan tri-oleates. This substitution makes Twee 85 more lipophilic with an HLB of 11.0 compared to Twee 80, which has an HLB of 15.0 (Yuan et al., 2006).

Twee 80 is hydrophilic in nature because the hydroxyl groups on the sorbitan ring are replaced and substituted with bulky polyoxyethylenes groups (Yaghmur et al., 2004). This substitution makes Twee 80 more soluble in water, so it tends to form O/W emulsion (Jiao and Burgess, 2006). Figure 3.3 shows the ternary phase diagram of Twee 80/Sunflower Oil/Deionized Water. It was observed that homogeneous region was formed along the apex line of Twee 80 and Sunflower Oil. This is because Twee 80 formed larger O/W emulsion areas as compared to the other surfactants due to higher HLB value of 15.0 of Twee 80, which promoted the formation of O/W emulsion (Golemanov et al., 2006). The formation of a one phase region suggested that the surfactant mixtures were able to lower the surface tension between the aqueous phase and oil phase, hence facilitate the formation of emulsions having a milky appearance (Mat Hadzir et al., 2013). Even though Twee 80 is the best surfactant to form O/W emulsion in the system, the formation of two phases is still exists. The two-phase region was the largest region observed in the ternary phase diagram because it covered most of the water and Sunflower Oil apex line. This showed the instability and incompatibility in the emulsion system.

3.4 Phase Behavior of Twee 85/VCO/Water

Figure 3.4 shows the ternary phase diagram of Twee 85/VCO/Deionized Water. The HLB value of Twee 85 (11.0) was lower as compared to Twee 80 (15.0). The regions that appeared in this ternary phase diagram were two-phase (T), homogeneous (H) and isotropic (L) regions. Isotropic region appeared at the percentage of Twee 85 at 3% to 88% and the deionized water at 12% to 97%. The homogeneous region, milky emulsion system was found along the apex line of Twee 85 and VCO. However, increasing the percentage of deionized water caused the formation of two-phase region. Two-phase region was the largest region that was observed in the ternary phase diagram. The two phases regions also appeared when using Twee 80 as an emulsifier. In Twee 85, two hydroxyl groups of the polyethylene moiety of Twee 80 are substituted by two lipophilic oleate tail groups and form polyoxyethylenesorbitan tri-oleates. It makes Twee 85 more lipophilic with an HLB of 11.0 compared to Twee 80, which has an HLB of 15.0 (Yuan et al., 2006).
3.5 Phase Behavior of Tween 80/VCO/Water

Figure 3.5 shows the ternary phase diagram of Tween 80/VCO/Deionized Water. The HLB value of Tween 80 (15.0) was higher as compared to the HLB value of Tween 85 (11.0). Tween 80 is a water soluble surfactant (Withhayapanyaneton et al., 2008). The isotropic region was found along the apex line of Tween 80 at 10% to 100% and the deionized water at 0% to 90%. Isotropic regions appeared in both Tween 85/VCO/Deionized Water and Tween 80/VCO/Deionized Water system, meanwhile, in Tween 85/Sunflower Oil/Deionized Water and Tween 80/Sunflower Oil/Deionized Water there is no isotropic region found. The homogeneous region appeared at the top of the ternary phase diagram where the percentage of Tween 80 was from 52% to 95% and the percentage of deionized water was from 5% to 58%. The formation of a one phase region suggested that the surfactant mixtures were able to lower the surface tension between the aqueous phase and oil phase, hence facilitate the formation of emulsions having a milky appearance (Mat Hadzire et al., 2013). Two-phase region was the largest region that was observed in the ternary phase diagram.

3.6 Phase Behavior of Tween 85/Olive Oil/Water

Figure 3.6 depicts the ternary phase diagram of Tween 85/Olive Oil/Deionized Water. The homogeneous region, milky emulsion system was formed along the apex line of Tween 85 and deionized water. However, increasing the percentage of deionized water caused the formation of two-phase region. The two-phase region was found at the water-rich corner in the Tween 85/Olive Oil/Deionized Water system. It covers the most of the water and Olive Oil. There is no isotropic region found in Tween 85/Olive Oil/Deionized Water system. In Tween 85, two hydroxyl groups of the polyethylene moiety of Tween 80 are substituted by two lipophilic oleate tail groups and form polyoxyethylenesorbitan tri-oleates. This substitution makes Tween 85 more lipophilic with an HLB of 11.0 compared to Tween 80, which has an HLB of 15.0 (Yuan et al., 2006).

3.7 Phase Behavior of Tween 80/Olive Oil/Water

Figure 3.7 shows the ternary phase diagram of Tween 80/Olive Oil/Deionized Water. The regions that appeared in this ternary phase diagram were two-phase (T), homogeneous (H) and isotropic (L) regions. Tween 80 is hydrophilic in nature because the hydroxyl groups on the sorbitan ring are replaced and substituted with bulky polyoxyethylene groups (Yaghmuret et al., 2004). This substitution makes Tween 80 more soluble in water, so it tends to form O/W emulsion (Jiao and Burgess, 2006). In Tween 80/Olive Oil/Deionized Water system, isotropic region appeared at the percentage of Tween 80 at 10% to 88% and the deionized water at 12% to 90%, meanwhile, there is no isotropic region appeared in Tween 85/Olive Oil/Deionized Water system. This is due to the stabilization of oil-in-water emulsions. The homogeneous region, milky emulsion system was found along the apex line of Tween 80 and Olive Oil. Two-phase region was the largest region that was observed in the ternary phase diagram that showed the instability and incompatibility in the emulsion system.
The HLB numbers (10-15) are detergents, and surfactants with HLB numbers between 7 and 9 are suitable for wetting agents (Griffin, 1949). According to Griffin’s theory (1954), to select a surfactant properly for any application, one must have the optimal HLB value and the correct chemical group. Thus, the best oil used in this study is VCO and the best surfactant used is Tween 80 which the HLB value is 15.0. In the ternary phase diagram of Tween 80/VCO/deionized water, Tween 80/VCO/deionized water and Tween 80/Olive Oil/deionized water, the presence of one phase region (homogeneous and isotropic) was observed. This showed that higher HLB value of the surfactant gave larger homogeneous and isotropic region in ternary phase diagram (AbdGanie et al., 2010). A high HLB value generally indicates good surfactant solubility in water, while a low HLB value indicates a high relative affinity for the oil phase.

**CONCLUSIONS**

The study of vegetable oils behavior with surfactant in emulsion system has potential applications, usefulness and advantages in industry. They act as deliver system by various routes of administration, as well as in food, cosmetics, and pharmaceuticals and so on. Thus, these vegetable oils behavior is important in development of emulsion-based products and could be applied in many industries.

**REFERENCES**


Figure 3.5: Ternary Phase Diagram of Tween 80/VCO/Deionized Water. (L-Isotropic region; H - Homogeneous region; T - Two phase region)

Figure 3.6: Ternary Phase Diagram of Tween 85/Olive Oil/Deionized Water. (H- Homogenous region; T - Two phase region)

Figure 3.7: Ternary Phase Diagram of Tween 80/Olive Oil/Deionized Water. (L- Isotropic region; H - Homogeneous region; T - Two phase region)

Results from this study demonstrated the importance of selecting oil and a surfactant with proper HLB for specific oil, as well as the type of surfactant. In order to determine the suitable surfactant that could be used for several types of emulsions or not, the HLB number could be used as a reference. In general, surfactants with lower HLB numbers (4-6) are mostly used as emulsifiers (water-in-oil), while those with higher HLB numbers (10-15) are detergents, and surfactants with HLB numbers between 7 and 9 are suitable for wetting agents (Griffin, 1949). According to Griffin’s theory (1954), to select a surfactant properly for any application, one must have the optimal HLB value and the correct chemical group. Thus, the best oil used in this study is VCO and the best surfactant used is Tween 80 which the HLB value is 15.0. In the ternary phase diagram of Tween 80/VCO/deionized water, Tween 80/VCO/deionized water and Tween 80/Olive Oil/deionized water, the presence of one phase region (homogeneous and isotropic) was observed. This showed that higher HLB value of the surfactant gave larger homogeneous and isotropic region in ternary phase diagram (AbdGanie et al., 2010). A high HLB value generally indicates good surfactant solubility in water, while a low HLB value indicates a high relative affinity for the oil phase.

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The study of vegetable oils behavior with surfactant in emulsion system has potential applications, usefulness and advantages in industry. They act as deliver system by various routes of administration, as well as in food, cosmetics, and pharmaceuticals and so on. Thus, these vegetable oils behavior is important in development of emulsion-based products and could be applied in many industries.

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