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Development of Emulsification containing Natural Colorant from Local Plant (Roselle)

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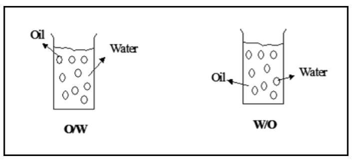
Graphical abstract



Abstract

Application of water-in-oil emulsion in the formulation of color cosmetic is very common. However using natural water-soluble pigments as cosmetic colorant is not general in the cosmetic industry although we knew the side effect from using synthetic colorant. This study concerns one of the most important problems in using natural pigment which was to obtain the lip moisturizer formulation and to determine emulsion stability. Roselle is a local potential pigment source which is added into castor oil as the oil phase with the aid of Span 80 and Tween 85 as emulsifier. Several emulsions were prepared using different proportions of emulsifiers and the most suitable hydrophilic lipophilic balance (HLB) value for color emulsions was determined to continue with the finding of stable formulation. In addition, stable formulation was determined and emulsions stability were studied during the observation period. Emulsions were analyzed by using viscosity measurement, in order to determine a suitable formulation of emulsion. The formulation of emulsion was stabilized under HLB value of 5.3 which is 75% of Span 80 and 25 % of Tween 85.

Keywords: Roselle; emulsion; emulsifier; cosmetic colorant; homogenization

1.0 INTRODUCTION

Natural ingredients are nowadays highly demanded whether for food, drugs and personal care products. The U.S. Department of Agriculture (USDA) permits the use of term "natural" on the labeling of food if the products contain no artificial flavor or flavoring, coloring ingredients, chemical preservative, or any other artificial or synthetic ingredient. Unlike food products, there are no finished cosmetic products that occur in nature. It is due to the commercialized products using synthetic color additives.

Synthetic colorant is undesirable because of its chemical effect. In contrast, natural colorant gives more benefit within its highly nutritional content. Natural colorant is produced from the natural pigment of plant such as anthocyanin, carotenoid and betalain. In recent years, most of the colorants in food used are from the natural colorant because of its nutritional advantages however it is rare or even no such cosmetic products commercially available yet.

Pigments extracted from plant roselle are widely used as natural red color in food. This natural plant pigments, named anthocyanin is in bright purple-red. It brings benefits in contrast to synthetic pigments. For example roselle contain active compound alpha-hydroxy acid (AHA), ascorbic acid, citric acid and other fruits acid. AHA has been proved to be effective in minimizing the wrinkles and fine lines [1] which is the flavor of ladies with deep vertical lines and dryness of lips. Other major benefit of natural roselle pigments is containing the antioxidant that would help in slowing down the ageing process for cosmetic purposes [2].

Emulsions have found numerous applications in many personal care and cosmetic products. Emulsion system consists of an immiscible liquid (internal) phase finely dispersed in another liquid (external) phase; for example oil dispersed in water (o/w emulsion) or water dispersed in oil (w/o emulsion). Color cosmetic, for example lipsticks are w/o emulsion applied products for moisturizing purpose. Figure 1 shows the basic explanation of o/w and w/o emulsion structures. Basically, the molecules contact of o/w and w/o emulsions are almost the same in terms of its molecule movement behaviour.

Fluids w/o emulsions generally present low stability because of the high mobility of water droplets, which can easily sediment, flocculate or coalesce [3]. Emulsions are unstable system due to an excess surface free energy where stability implies no tendency on structural changes [4]. In order to overcome the breakdown of two immiscible liquids, emulsifier/s is used to maintain stable emulsion properties for a period of time. It means that both liquids are not separated after the agitation has stopped.

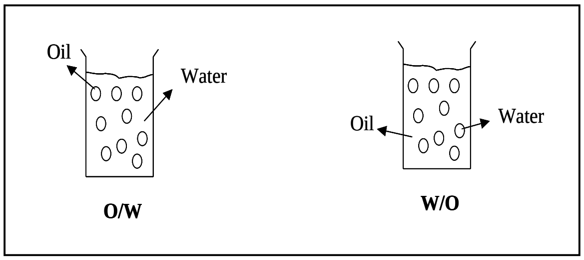


Figure 1 The schematic representation of two types of emulsion

Stability is a very important parameter to know the applicability of an emulsion to produce more complex systems. Because the emulsions studied in this work are wanted to be used in the formulation of cosmetic products, a relatively high stability is needed to get a product of invariable quality during the required time (transport, storage, etc.) [5].

The objective of this work was to obtain the formulation of w/o color emulsions with Span 80 and Tween 85 as a potential practical application to prepare color cosmetics. Appearances of emulsions were observed and characterizations of emulsions were performed by viscosity measurement, which is a reliable measure of emulsion stability [6].

■2.0 EXPERIMENTAL

2.1 Materials

Roselle flowers were obtained from Negeri Sembilan, Malaysia. Calyces of roselle were collected to be dried in an oven at 35°C. The seeds were removed before the drying process. Dried roselles were maintained in glass jars in refrigerated conditions (3±1°C) until used for extraction. In order to make the emulsion, Span 80 and Tween 85 were used as emulsifiers. In addition, castor oil was used as the oil medium for the w/o emulsion.

2.2 Extraction of Roselle

Water-based pigments were extracted from calyces of roselle by solvent extraction method adapted from Salazar-Gonzalez *et al*. (2012) [7] with modifications. Dried roselle calyces were grinded using blender before extraction. Grinded roselle (20 g) was placed in glass beakers, and 200 ml of the extracting agent, Ethanol 96% was added. The beakers were protected from light with aluminium foil and the mixture was stirred for 24 hours. The supernatant was filtered through Whatman paper No.2 and placed in spherical flasks. The extract was further treated using rotary evaporator to obtain a pure roselle extracts.

2.3 Preparation of Emulsions

The preparation of color emulsions was adapted from Rukmini *et al*. (2012) [8] with modifications. The emulsion systems were prepared with surfactants mixture consisting of Span 80 and Tween 85 to achieve predetermined HLB values of 4.3, 4.6, 5.3, 6.0 and 6.3 through calculation using the equation shown below.

[% of Span 80 × HLB value of Span 80 (4.3)] + [% of Tween 85 × HLB value of Twenn 85 (11.0)] / 100% = Final HLB value

Briefly, roselle and a combination of surfactant mixtures with combinations ratio of 1:1, 1:2, 1:3, 1:4, 1:5 and 1:6(v/v) were prepared and mixed using homogenizer at a lower speed. After ten minutes, castor oil was added dropwise while homogenize at high speed. The formation of emulsion system was characterized by the presence of clear and transparent solutions. These emulsions were allowed to equilibrate for at least 24 hours before further measurements on stability.

2.4 Stability Test of Emulsions

The stability of emulsions during storage at room temperature was examined. All of the emulsion samples were stored in 10ml glass sample bottles. Classical method of stability tests undergone in this study are viscosity measurements using Brookfield Viscometer model DV-1 within the period of 14 days in different temperatures of 4°C, room temperature of 27°C and 60°C . Readings were taken after one day trial period, 7 days and 14 days.

■3.0 RESULTS AND DISCUSSION

3.1 Suitable HLB Values for Color Emulsions

The roselle-in-castor oil emulsions in this study were prepared by using surfactants combination of Span 80 and Tween 85 to achieve a variety of HLB values that are suitable for water-in-oil emulsion. The concept on which this procedure is based, the hydrophile–lipophile balance (HLB), is that the molecule of any emulsifier, or indeed of any surface-active material, contains both hydrophobic and hydrophilic groups, and the ratio of their respective weight percentages should influence emulsification behavior [9].

The HLB value indicates how the surfactant will behave in a solution. Span 80 have a HLB value of 4.3 which is a relatively low HLB emulsifier with lipophilic properties while tween 85 have a HLB value of 11 which is a relatively high HLB emulsifier with hydrophilic properties. The combinations of two or more emulsifiers are able to form a stable emulsion effectively [3, 8].

In this study, the combinations of emulsifiers were mixed to achieve predetermined HLB values of 4.3, 4.6, 5.3, 6.0 and 6.3 as described before. It was found that the HLB value that make the emulsion most stable was HLB 5.3 which was composed of 75% of Span 80 and 25% of Tween 85. The transparent appearance of the w/o emulsion using surfactants having the HLB value of 5.3 was selected for further experiments.

3.2 Optima Formula of Color Emulsions

The emulsion system can be formulated by appropriate proportions of water, surfactants and oil. In order to obtain the suitable formulation for roselle-in-castor oil emulsion, the various combination of roselle/surfactants/castor oil at different ratios were prepared (Table 1).

Transparent w/o emulsions were formed when the ratios of water to surfactants were of at least 1:4 and the ratio of water/surfactant mixture to castor oil was 1:3 which means the emulsion system consist of 75% of castor oil. the castor oil was added as the continuous phase, clear appearance of the emulsions were obtained as shown in formulation 4, 5 and 6. However, when the castor oil was added at a higher ratio as 1: 4, transparent w/o emulsion was not achievable as indicated by formula 7.

The formula of 1, 2 and 3 could not achieve emulsions system because of insufficient surfactants in the systems. According to Patel *et al*. [9], one of the important factors to be considered during emulsion preparation is that surfactants must be high enough to provide the number of surfactant molecules needed to stabilize the droplets in emulsions. When the surfactant concentration is decreased, the droplet size logically increases [10]. Consequently, the transparent systems obtained from the formulae of 4, 5 and 6 were selected and used for further experiment and were coded as A, B and C.

Table 1 Evaluation of roselle-in-castor oil emulsion formulation

Formula	Ratio of roselle/ surfactants (w/s)	Ratio of (w/s): castor oil	Appearance		
			24 hours	7 days	14 days
1	1:1	1:3	c	c, s	c, s
2	1:2	1:3	c	c, s	c, s
3	1:3	1:3	c	c, s	c, s
4	1:4	1:3	t	t	t
5	1:5	1:3	t	t	t
6	1:6	1:3	t	t	t
7	1:5	1:4	c	c, s	c, s

c: cloudy; t: transparent; s: separate

3.3 Stability Tests

Formulation A, B and C are continued for stability analysis. Table 2 indicates the viscosity of three emulsion formulations. Viscosity can be used to detect the emulsion structural changes due to it depends largely on the emulsion structure, i.e., the type and shape of aggregates, concentration and interaction between dispersed particles [11, 12].

In the study of Ilia Anisa and Nour (2010) [13] have proven the changing of droplet diameter was depends on the viscosity and the behavior of emulsion. In another study also showed that the droplet size is depended on the phase fractions which as the droplet size increase, the competing rates of coalescence and breakage increase [14].

Table 2 Viscosity measurement of emulsions

Formula	Viscosity (cp)		
	24 hours	7 days	14 days
A	1007.5	1007.5	999.6
B	1005.6	1002.3	998.4
C	999.5	999.2	997.5

■4.0 CONCLUSION

As a conclusion, the unstable behavior of natural colorant could be overcome by proposing the development of application of emulsion. The formulation of emulsion was stable under HLB value 5.3 which is 75% of Span 80 and 25% of Tween 85. While ratio of water/ combination surfactant should be at least 1:4 and ratio of (w/s) to oil is 1:3, which is the 75% in system. High value of viscosity is in need especially for the purpose of cosmetic formulation.

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References

[1] Masri, R., R. Ismail and S. Ahmad. 2002. Palm-based Cosmetic Products with Roselle Extract. *MPOB Information Series.* 156–160.

[2] Batista, A. P., A. Raymundo, I. Sousa and J. Empis. 2006. Rheological Characterization of Coloured Oil-in-Water Food Emulsions with Lutein and Phycocyanin Added to the Oil and Aqueous Phases. *Food Hydrocolloids.* 20(1): 44–52.

[3] Ushikubo F. Y. and R. L. Cunha. 2012. Stability Mechanisms of Liquid Water-in-Oil Emulsions. *Food Hyfrocolloids.* 1–9.

[4] Pittia, P., A. Gambi and C. R. Lerici. 1997. Hygrometric Measurements for the Evaluation of the Stability of Model Food Emulsions. *Food Research International.* 30(3–4): 177–184.

[5] Andres, L. Marquez, Gonzalo, G. Palazolo and Jorge, R. Wagner. 2007. Water in Oil and Double Emulsions Prepared with Spans: Microstructure, Stability and Rheology. *Colloid Polym Sci.* 285: 1119–1128.

[6] Chen, G. and D. Tao. 2005. An Experimental Study of Stability of Oil-Water Emulsion. *Fuel Processing Technology.* 86: 488–508.

[7] Slazar-Gonzalez C., F. T. Vergara-Balderas and A. E. Ortega-Regules. 2012. Antioxidant Properties and Color of Hibiscus Sabdariffa Extracts. *Ciencia e Investigacion Agraria.* 39(1): 79–90.

[8] Rukmini, A., S. Raharjo, P. Hastuti and S. Supriyadi. 2012. Formulation and Stability of Water-in-Virgin Coconut Oil Microemulsion Using Ternary Food Grade Nonionic Surfactants. *International Food Research Journal.* 19(1): 259–264.

[9] Patel, M. R., R. B. Patel, J. R. Parikh, K. K. Bhatt and A. J. Kundawala. 2009. Microemulsion : As Novel Drug Delivery Vehicle.

[10] Roland I., G. Piel, L. Delattre and B. Evrard. 2003. Systematic Characterization of Oil-in-Water Emulsions for Formulated Design. *International Journal of Pharmaceutics.* 263: 85–94.

[11] Lim, W. H. 2006. Phase Diagram, Viscosity and Conductivity of A-Sulfonate Methyl Esters Derived From Palm Stearin/1-Butanol/Alkane/Water Systems. *Journal of Surfactants and Detergents.* 9(4): 349–355.

[12] Garti, N., A. Spernath, A. Aserin and R. Lutz. 2005. Nano-sized Self-assemblies of Nonionic Surfactants as Solubilization Reservoirs and Microreactors for Food Systems. *Soft Matter.* 1: 206–218.

[13] Ilia Ansia, A. N. and A. H. Nour. 2010. Affect of Viscosity and Droplet Diameter on Water-in-Oil (W/O) Emulsions: An Experimental Study. *World Academy of Science, Engineering and Technology.* 38: 691–694.

[14] Rueger, P. E. and R. V. Calabrese. 2012. The Effect of Phase Fraction on Drop Size Distribution in a High Shear Mixer with a Viscous Continuous Phase. *14th European Conference on Mixing Warszawa.*