

Winsor Phase Diagram of a Colloidal System from the Mixture of Water, Eugenol, and Tween 20

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ABSTRACT

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One factor that influenced colloidal structure is the composition of water, oil, and surfactant in the emulsions. This study aims to build a Winsor phase diagram of a mixture of water, eugenol, and Tween 20 and understand the physical differences in a range of micellar structures from the different compositions of the combinations. There were eleven samples with varying compositions of water, eugenol, and tween 20, and then were mixed and observed in parameters such as phase, appearance, and consistency. The results showed that the emulsion's compositions ingredients affected the character of the final emulsion. There were three categories of emulsions as described in the Winsor phase diagram. Water in oil (w/o) microemulsion was formed in the higher oil composition. In contrast, the lower oil content was macroemulsion/ coarse emulsion. The balance of oil and water composition was categorized as a bicontinuous microemulsion. This diagram will further help for constructing the suitable emulsion category for specific purposes.

Keywords: colloidal structure, emulsion, Winsor diagram, Phase equilibrium diagram, microemulsion

1. INTRODUCTION

The micellar system is the colloidal system that consists of a mixture of water, oil, and an amphiphile surfactant[1], [2]. The amphiphilic surfactant is added to increase its miscibility[3]. The amphiphile surfactant is either not charged (i.e., is nonionic) or consists of an anionic (charged) headgroup[4]. The phase diagram of the ternary micellar system is represented by a triangle (A: water, B: oil, and C: surfactant). Their characteristic mixed-phase describes the three binary methods A-B, A-C, and B-C (1-phase) and phase-separated (2-phase) regions[5]. The oil surfactant (B-C) binary mixture is characterized by an upper critical solution temperature (UCST); i.e., its phase separates upon cooling. The water-oil (A-B) binary mixture is characterized by UCST but is mostly phase-separated.

The phase diagram for the water-surfactant (A-C) binary solution is more complex and characterized by a UCST behavior at low temperatures and a closed-loop immiscibility island at high temperatures; i.e., it phase separates both upon cooling upon heating. The closed-loop is due to the breaking of hydrogen bonds upon heating and shows up for strong amphiphile surfactants[6]. This character of a phase diagram of the micellar system is essential for some colloidal mixture containing oil, water, and emulsifier in the food, pharmaceutical, and oil industries[7], [8].

In 1948, Winsor found four-phase equilibrium systems from mixtures of water, oil, and an amphiphilic compound, later called Winsor I, II, III, and IV systems. The Winsor I system is a lower dispersion in equilibrium with an excess oil upper phase. In contrast, the Winsor II system is an upper dispersion

in equilibrium with an excess water lower phase. The Winsor III system has a three-phase structure, with a middle distribution in equilibrium with extra oil upper and excess water lower phases. The last one, Winsor IV, is a macroscopically single-phase dispersion [9], [10]. Most surfactant resides in the shaded area. In a three-phase system, the middle-phase microencapsulation (M) is in equilibrium with both excess oil (O) and water (W)[10]

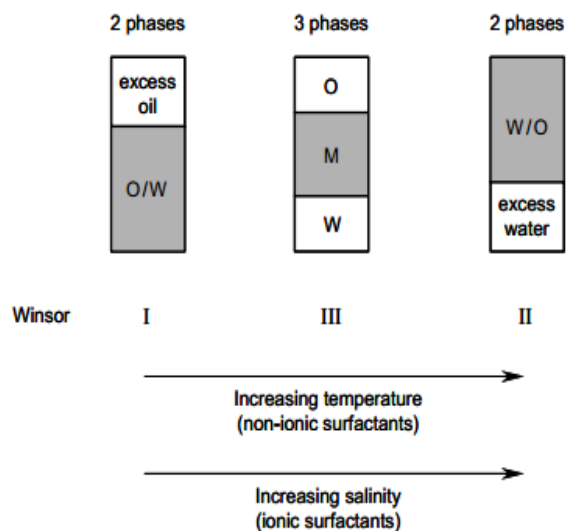


Fig 1. Winsor classification and phase sequence of microemulsions encountered as temperature or salinity are scanned for nonionic and ionic surfactant, respectively.

Microemulsion can be defined as a thermodynamically stable dispersion macroscopically monophasic, fluid, and transparent. Microemulsions can consist of isotropic mixtures that are combined with oil, water, a surfactant, and often combined with a cosurfactant. It is stabilized by an interfacial film composed of the surfactant, forming droplets with diameters ranging from 5 to 100 nm. They are formed spontaneously when the aqueous phase, oil, and surfactant come into contact and have the ability to combine enormous quantities of two immiscible liquids in a single homogeneous phase[11], [12].

This study aims to build a Winsor phase diagram from a mixture of water, eugenol, and Tween 20 and understand the physical differences in a range of micellar structures from the different compositions of the combinations.

2. MATERIALS AND METHODS

2.1 Materials

Tween 20 (Polyethylene glycol sorbitan monolaurate, polyoxyethylenes sorbitan monolaurate) as a surfactant, eugenol as an essential oil.

2.2 Preparation of micellar system

Eleven different solutions were prepared by the different ratios of water:oil: surfactant that described in table 1. The solutions were then mixed by using vortex for two minutes and observed by the following parameter:(a) Phase: by monitoring how many layers and separation, (b) Appearance / Turbidity, and (c) Consistency / Viscosity

Table 1. Different ratio of water: Eugenol: Tween 20 combinations

Run.	Water (%w/w)	Eugenol (%w/w)	Tween 20 (%w/w)
1.	33.33	33.33	33.33
2.	50	50	0
3.	0	50	50
4.	50	0	50
5.	25	0	75
6.	60	10	30
7.	30	60	10
8.	10	30	60
9.	60	30	10
10.	10	60	30
11.	30	10	60

3. RESULTS AND DISCUSSION

In this experiment, the composition of the solution is based on the composition in **Table 1**. The emulsion result showed in **Fig 2**. It can be seen that there were emulsion results in different phases, appearance, and consistency. Based on the various physical characteristic of emulsion, we could categorize those emulsions as three areas (condition).

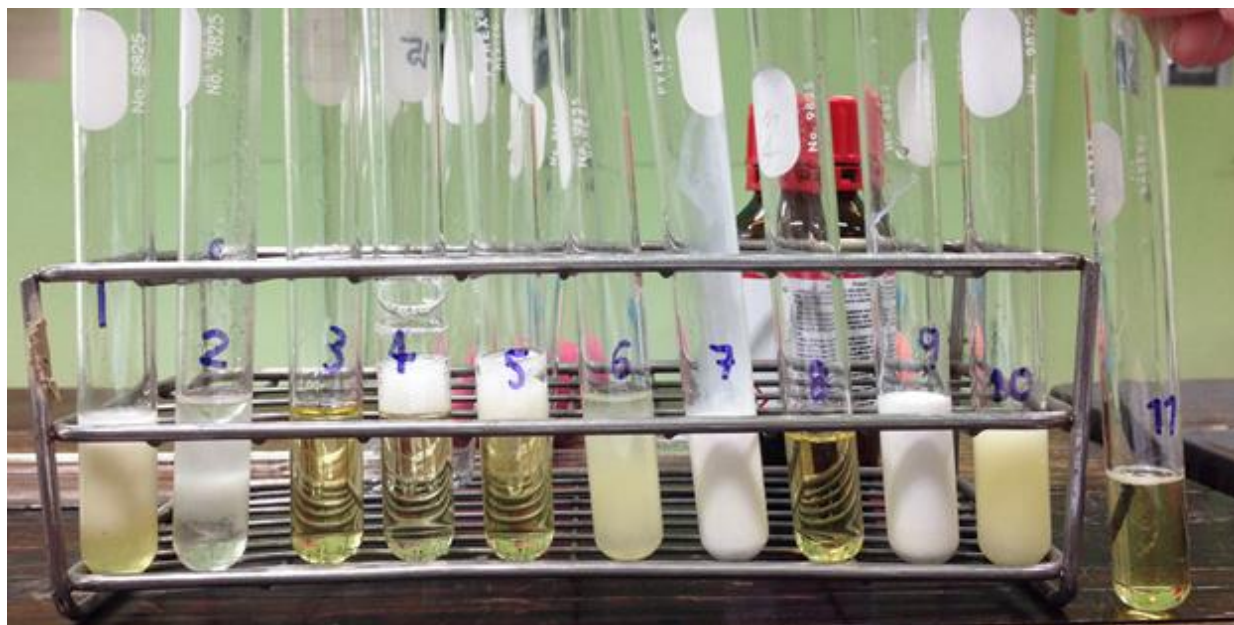


Fig 2. The photograph of emulsion results from varying concentrations of water, eugenol and tween 20

The feature of the emulsions categorized as w/o microemulsion is described in Table 2. Table 3 shows the characteristic of emulsion in the bicontinuous microemulsion. Whereas table 4 shows the feature of coarse emulsions/ macroemulsions. Based on those characteristics and the type of emulsion, can be constructed Winsor phase as shown in Fig 3.

Tables 2 and 3 showed the physical characteristic of the microemulsion. It defined emulsion in this category because it has several aspects such as optically transparent (isotropic), may be single or multiple phases, and thermodynamically stable. An emulsion is defined as a transparent, isotropic system composed of a relatively large fraction of surfactant and co-surfactants compared to water and oil phases. In such systems, dispersed phase droplet does not refract light; hence the dispersed phase globules appear invisible to the naked eye. Globule size ranged below 200nm, so such a system became transparent. Microemulsion has been shown to possess ultra-low interfacial tension (sometimes negative) as it can accommodate a significant fraction of co-surfactant/surfactant mixture at the interface, thus thermodynamically stable [12], [13]

Table 4 shows the emulsions that were categorized as macroemulsions. Macroemulsion is a coarse emulsion characterized by thermodynamically

unstable, direct oil/water contact at the interface, multiple phases only, and cloudy colloidal systems [9]. The Winsor phase diagram 3 component mixture of water, eugenol, and tween 20 in Fig 3 showed the physical characteristic of emulsions. We defined that diagram into three areas. The pink area was designated as w/o microemulsion resulting from stable emulsion (Table 2). This area was characterized as one phase and apparent appearances due to micellar formation that resulted from the excess amount of surfactant. Samples no 4 and 5 showed gas bubbles in the top of emulsions due to the higher extra amount of surfactant.

Due to multiple phases and turbidity, the green area was defined as bicontinuous microemulsion (Table 3). Sample no one was categorized as Winsor III, which formed three layers as shown in Fig 1. The top layer was oil, the middle was microemulsion, and the bottom was water. Sample no 6 was oil in water microemulsion, and the top layer was excess of water (Winsor I). Whereas sample no 10 is water in water emulsion, the top layer was excess of oil (Winsor II).

Macroemulsion was defined in the blue area. Those emulsions that were not stable made multiples layers and milky appearance (Table 4). Those emulsions would be separated into multiple phases due to the lack of surfactant.

Table 2 The physical characteristics emulsions in w/o microemulsions .

vial number	Compositions (%)			phases	Appearance	Consistency	Condition	Picture
	Water	Eugenol (oil)	Surfactant (tween 20)					
3	0	50	50	1 phase	Clear /Not turbid	Not viscos	Micellar (microemulsion)	Sample no 3
8	10	30	60	1 phase	Clear /Not turbid	Not viscos	Micellar (microemulsion)	Sample no 8
11	30	10	60	1 phase	Clear /Not turbid	Not viscos	Micellar (microemulsion)	Sample no 11
5	25	0	75	1 phase	Clear /Not turbid	viscos ++	Micellar (microemulsion)	Sample no 5
4	50	0	50	1 phase	Clear/Not turbid	Viscos +	Micellar	Sample no 4

Table 3. The physical characteristics emulsion in bicontinuous microemulsion.

vial number	Compositions (%)			phases	Appearance	Consistency	Condition	Picture
	Water	Eugenol (oil)	Surfactant (tween 20)					
1	33.33	33.33	33.33	3 phases	separated into 3 layer, the center phase is turbid, bottom and top are clear	only the centre layer is viscos	Bicontinuous microemulsion	Sample no 1
6	60	10	30	2 phases	separated into 2 layer, bottom is turbid, top clear	2 layers. Top layer is excess water, the bottos is microemulsion	o/w emulsion with excess water	Sample no 6
10	10	60	30	2 phase	Turbid, top layer is excess oil	Not viscos	w/o microemulsion	Sample no 10

Table 4. The physical characteristic emulsion in macroemulsion/ coarse emulsion

vial number	Compositions (%)			phases	Appearance	Consistency	Condition	Picture
	Water	Eugenol (oil)	Surfactant (tween 20)					
7	30	60	10	2 phases	Turbid +++	not viscos	Macroemulsion (not stable)	Sample no 7
9	60	30	10	2 phases	Turbid +++, in the bottom just a bit excess water	not viscos	Macroemulsion (not stable)	Sample no 9
2	50	50	0	3 phases	in the center layer is turbid++, top and bottom layer excess water (clear)	not viscos	Macroemulsion (not stable)	Sample no 2

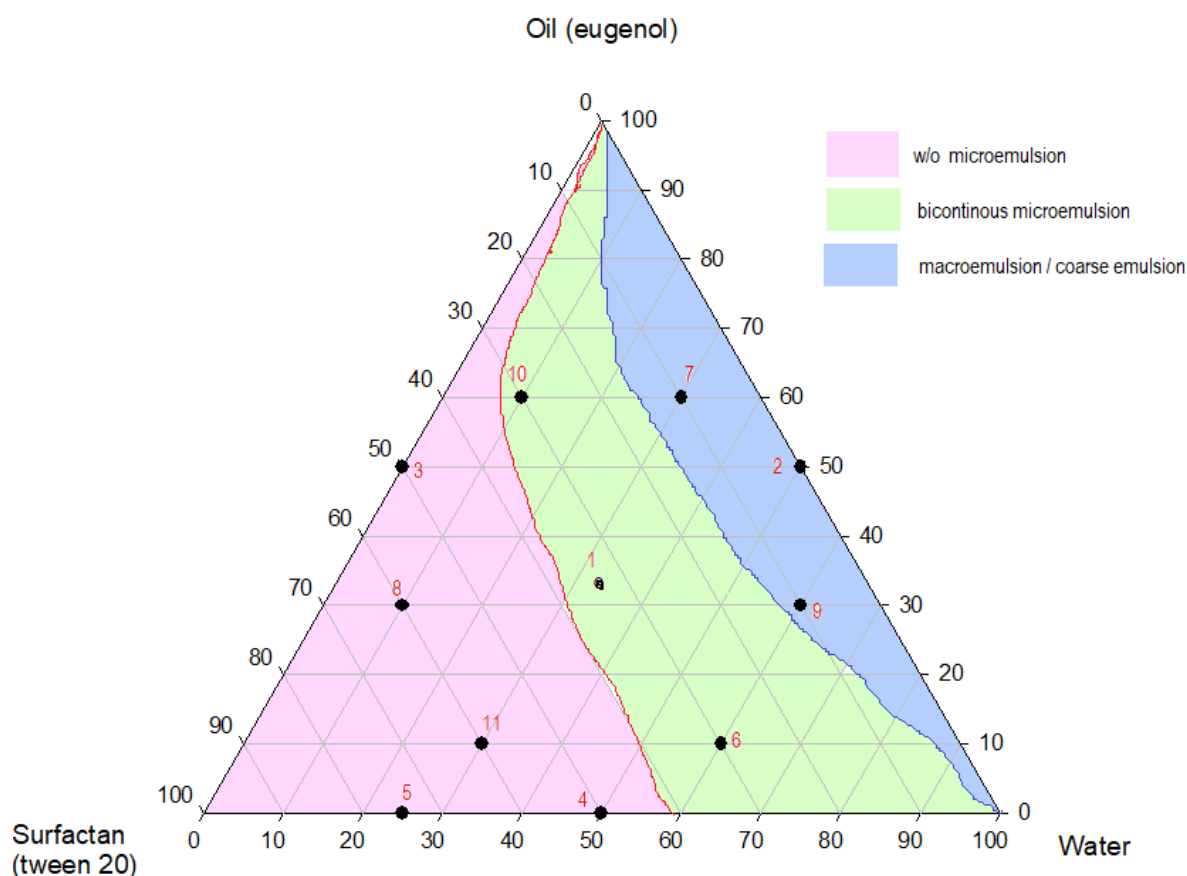


Fig 1. Winsor phase 3- components mixture of water: Eugenol: tween 20

The red number represents the vials numbers of emulsion samples

4. CONCLUSION

One factor that influenced colloidal structure is the composition of water, oil, and surfactant in the emulsions. The macroemulsion is a coarse emulsion that is not stable compared to the microemulsion due to a lack of surfactant.

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