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Effect of Dispersed Phase Volume Fraction on Physical Stability of Oil-in-Water Emulsion in The Presence of Gum Tragacanth

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Abstract

Background: The dispersed phase volume fraction plays key role in oil-in-water emulsion stability by different mechanisms like partial flocculation, depletion flocculation and bridging flocculation. Since the mentioned mechanism is affected by emulsifier concentration presented in system, the major aim of this study was to investigate the effect of dispersed phase volume fraction on physical stability of oil-in-water emulsion in the presence of gum tragacanth.

Methods: The effect of oil concentration on the stability of oil-in-water emulsion systems containing 0.5% w/w gum tragacanth is investigated by considering rheological properties and droplet size distribution. Emulsion stability was monitored by measuring the extent of gravitational phase separation. Rheological properties and droplet size distribution were measured by rheometer and static laser diffraction immediately after preparation at 25 °C. All treatments were performed three times and the data were analyzed using SPSS 16.0.

Results: Oil concentration enhancement from 10% to 30% w/w decreased emulsion stability. Increasing the oil concentration enhances consistency coefficient from 2.01 to 5.73 pa.sⁿ but decreases flow behavior index. Considering the particle size distribution revealed that number mean diameter increase significantly by oil concentration addition.

Conclusion: Emulsion stability index decreased by increasing dispersed phase volume fraction. Considering rheological properties and droplet size distribution, it seems that oil concentration addition however, makes the system more structured but it is unable to create a stable system since the emulsifier is insufficient to protect oil droplets from aggregation.

Introduction

Emulsions are colloidal systems composed of two liquids; usually oil and water, which droplets of one liquid is dispersed in another one, named dispersed and continuous phase, respectively (McClements, 2005). According to the location of the oil and water phases within the system, emulsions are categorized in terms of oil-in-water and water-in-oil emulsion (Amid and Mirhosseini, 2013). Oil-in-water emulsion (O/W) consists of oil droplets in an aqueous phase and water-in-oil emulsion (W/O) composed of water drop-

lets dispersed in an oil phase (Mollakhalili Meybodi and Mohammadifar, 2014).

Oil-in-water emulsions are used in food and pharmaceutical industries to fortify manufactured materials by lipophilic, healthy ingredients (Achouri et al., 2012). They are thermodynamically unstable systems that physically break mainly due to creaming, flocculation, coalescence, gravitational separation, phase inversion and ostwald ripening mechanisms (Mollakhalili Meybodi et al., 2014). However, emulsions are thermodynamically unstable systems but it is

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possible to form kinetically stable one using components which called emulsifier and thickening agents (Dickinson, 2009). Separately, emulsifiers reduce oil-water interfacial tension by creating a protective layer around oil droplets. Thickening agent increases the continuous phase viscosity and will reduce oil droplets upward movement and their aggregation according to stocks law.

Gum tragacanth (GT) which is called bifunctional emulsifier will enhance oil-in-water emulsion stability as an emulsifier and thickening agent due to its two fractions namely bassorin as a water-swellable and tragacanthin as a water-soluble (Mohammadifar et al., 2006).

Dispersed phase concentration plays an important role in oil-in-water emulsion stability determination (Osborn and Akoh, 2004). However, there is some evidences that, increasing oil phase will enhance oil-in-water emulsion stability via weak flocculation of the particles (Mostefa and Tir, 2004; Thanasukarn et al., 2004). Other studies reveal that oil phase concentration role in oil-in-water emulsion stability determination is related to emulsifier concentration, since emulsifier concentration may enhance instability either via depletion flocculation (Wang et al., 2011); if it present at higher concentration to cover oil droplet; or bridging flocculation; if it isn't sufficient to cover all oil droplets (Chuah et al., 2014).

By regard to bifunctional properties of GT (Balaghi et al., 2011) and dispersed phase volume fraction role (Vanapalli et al., 2002) in oil-in-water emulsion stability determination, the aim of this study was to evaluate the effect of dispersed phase volume fraction on physical stability of oil-in-water emulsion in the presence of GT.

Materials and methods

Materials

Iranian GT (*Astragalus gossypinus*) was collected from plants growing in the central mountainous area of Isfahan province, Iran. The raw gum was grounded and sieved. Powdered gum with mesh size between 200 and 500 μ m was used in this study. Commercial sunflower oil from the same lot was purchased from a local market.

Preparation of gum dispersions

GT dispersions (0.5% w/w) were prepared by adding 0.5 g gum powder (200–500 μ m) to 89.5 ml distilled water, the mixture were kept on a magnetic stirrer at room temperature. Sodium azide (0.05% w/w) was added to prevent microbial growth. Gum was completely hydrated when its solution was stored at 4 $^{\circ}$ C for an overnight. These amounts were chosen based on previous studies (Farzi et al., 2013; (Mollakhalili Meybodi and Mohammadifar, 2014).

Production of oil/water emulsions

For the preparation of 10, 15, 20, 25, 30% (w/w) O/W emulsions 10, 15, 20, 25, 30 g sunflower oil were added, respectively to 90, 85, 80, 75, 70 g of the gum dispersions and homogenized for 15 min at 13500 xg (Mohammadifar et al., 2006) by Ultraturax (IKA T 25, Deutschland, Germany). In order to minimize temperature fluctuations, emulsions were coated in ice.

Emulsion stability index

Emulsion stability was monitored by measuring the extent of gravitational phase separation. Immediately after preparation, 10 ml of each emulsion was transferred into a cylindrical plastic tube, subsequently tightly sealed with a plastic cap to prevent evaporation and quiescently stored at 25 °C during 120 days. Relative rates of creaming were monitored by visual observation during set time intervals. The extent of phase separation was obtained by Emulsion Stability Index (ESI) using the following equation:

$$ESI\% = \frac{H_E - (H_C + H_S)}{H_E}$$
 (1)

Where H_E is the initial emulsion height, H_C is the height of the cream layer and H_S is the height of the sedimentation phase. Monitoring tests were performed in duplicate and the mean of the two individual trials were taken for analysis.

Emulsion droplet-size analysis

The particle size distribution of emulsions were measured immediately after preparation at room temperature (25 °C) by static laser diffraction with a Cilas 1090 particle size analyzer (Orleans, France) equipped with a 5 mw He/Ne (635 nm) laser beam. Emulsions were diluted by deionized water (1:100) to avoid multiple scattering effects. When droplets were fully dispersed and reached an obscuration of at least 6%, the measurement was started. The emulsions were stirred continuously during the measurements to ensure that they were homogeneous. Particle size measurements were reported as $D_{0.1}$, $D_{0.5}$ and $D_{0.9}$ on a volume basis that is the size of particle below which 10%, 50% and 90% of the sample particles lie, respectively. Other parameters were automatically calculated using the Fraunhofer theory by the software provided with the apparatus. The mean particle size (i.e., the De Broucker diameter), was calculated as follows:

$$D(3,4) = \frac{\sum n_i d_i^4}{\sum n_i d_i^8}$$
 (2)

$$D(1,0) = \frac{\sum n_i d_i^4}{\sum n_i}$$
(3)

Where n_i is the number of particles of class "i" and d_i is the diameter of class "i". Then span (The polydispersity of the size distribution) as a measure of the distribution width of particles in dispersion, was calculated using the following equation:

$$Span = \frac{d_{0.9} - d_{0.1}}{d_{0.5}} \tag{4}$$

It should be noted that, all the particle size measurements were monitored before any visual phase separation took place.

Rheological measurements of emulsions

Immediately after preparation of emulsions, steady state rheological measurements were performed with a Physica MCR 301 rheometer (Anton-Paar GmbH, Graz, Austria) using control shear rate mode. Rheological characteristics were acquired by concentric cylinder geometry with a radius ratio of 1.0846 at 25±0.01 °C. Rheological data were collected using Rheoplus software version 3.21 (Anton-Paar). The temperature was adjusted by a viscotherm VT2 with a Peltier system equipped with a fluid circulator (Anton Paar, GmbH) with an accuracy of ± 0.01 . In all experiments, the sample was left at rest in the measurement system for 1 min prior to each analysis to obtain thermal equilibrium. Indeed, emulsions were covered with solvent trap to prevent evaporation. The flow curves were obtained at 0.01-700 s⁻¹, and then rheological data were fitted with the power law model over mid-range shear rates.

Power law model:

$$\sigma(\tau) = m.\dot{\gamma}^n \tag{5}$$

According to power law model, k is the consistency coefficient (pa. S^n). $\dot{\gamma}$ is shear rate (1/s), and n is flow behavior index (dimensionless).

Statistical analysis

Analytical values are based on the mean and standard deviation of three replicates. Analysis of variance (ANOVA) was used for the data analysis (SPSS 16.0). When the F-values were significant (p<0.05) in ANOVA, Duncan's multiple-range test was used to compare treatment means. Statistical differences between samples were also calculated using student's t-test for selecting data. Values were considered to be significantly different if p<0.05.

Results

ESI for emulsion containing different oil concentration are presented in Fig. 1. As is shown, in all samples ESI de-

creased over the storage time. Results revealed that emulsion stability decreased by increasing oil concentrations (Fig. 1).

Flow curves of oil-in-water emulsions containing different oil concentration in the presence of 0.5% w/w GT are presented in Fig. 2. As it is illustrated, all samples showed shear thinning behavior or pseudoplastisity. In other words, apparent viscosity decreases for all systems by increasing the shear rate. However, the apparent viscosity amounts increase as oil concentration is increasing, too.

Power law parameters for all systems are presented in Table 1. They were calculated by equation 5. The high determination coefficients showed that the model was well fitted to the data. As shown, increasing the oil concentration increases the consistency coefficient (a) significantly (p<0.05) in a way that consistency index of systems containing 10% w/w oil concentration is 1.85 fold lower than 30% w/w oil containing emulsion. Flow behavior index, as an indicator of shear rate dependency of systems, is illustrated in Table 1. According to Table 1, the flow behavior index decreased as oil concentration increased.

In order to give information about the effect of oil concentration addition on the GTs ability to stabilize emulsion, diameter statistics including d (0.1), d (0.5), d (0.9), d (4,3), d (1,0) and span values for oil-in-water emulsions containing different oil concentration in the presence of 0.5% w/w GT are presented in Table 1. As illustrated in Table 2, addition of oil concentration changed particle size distribution. It is obvious that there are extensive differences between d (4,3) and d (1,0) values of all samples. Table 2 shows that increasing oil concentration from 10% to 30% will enhance number mean diameter by about 78% of its initial value. Results indicated that while the volume mean diameter and also polydispersity of particles were constant by increasing the oil concentration, a significant increase in d (1,0) and number mean diameter were observed.

Discussion

ESI was preliminarily monitored visually by measuring the height of distinct serum (translucent) layer formed at the bottom of emulsions after 0 (freshly prepared emulsions), 5, 15, 30, 60, 90, 120 days storage at ambient temperature. According to Stocks law, creaming index is an indirect indicator to provide information about the droplet aggregation extent in the emulsions (Tadros, 2004). In order to attain more insight into the emulsion destabilization mechanism, rheological properties and droplet size distribution of samples were determined in relation to oil concentration enhancement.

Steady state rheological property is a direct indicator of shear rates dependency of emulsion flow properties (Amid and Mirhosseini, 2013). Shear thinning behavior observed in systems may be due to deflocculation and/or deformation of the droplets on the disperse phase and elongation and alignment of flocs or water-swellable fraction of GT with shear rate field (Balaghi et al., 2010). By respect to Table 1, the consistency coefficient enhancement perceived by increasing oil concentration indicates that oil concentration increasing intensified emulsion structure via droplet interaction improvement (Dokić et al., 2012). Flow behavior index decreases by increasing oil concentration more than 20% w/w. It shows that higher oil concentration containing emulsions are more pseudoplasticity (Balaghi et al., 2010). In other words, they got more dependants to shear rate than those containing lower oil concentration.

Droplet size distribution is an important parameter in determining emulsions characteristic such as the rheology (Asano and Sotoyama, 1999), the emulsion stability (Karimi and Mohammadifar, 2014) and stabilizers capability (Gallegos et al., 2009). By regard to the Stocks law, the rate of phase separation in an emulsion is proportional to the square of its dispersed phase radius. GT was placed at oil/water interface in a manner to delay adverse interaction and reduce particle size to a large amount (Farzi et al., 2013).

It is worthy to note that d (0.1), d (0.5), d (0.9), d (4,3) parameters are sensitive to particles volume presented in system and d (1,0) is in direct correlation with number of parti-

cles in distribution (Taherian et al., 2006). Span value is sensitive to volume of particles presented in systems leading to evaluate the effect of oil concentration on polydispersity of large polysaccharide particles and emulsion droplets or aggregates (Gallegos et al., 2009).

System particles are individual oil droplets, oil droplet aggregates, water soluble (tragacanthin) and water-swellable (bassorin) fractions of GT. The fractions are able to increase oil-in-water emulsion stability either directly or via reducing oil droplet aggregation process.

Considering the constant values of volume mean diameter by increasing oil concentration, the increase in number mean diameter of systems containing higher oil concentration can be related to the association of small oil droplet.

Conclusion

The results indicated that oil-in-water emulsion stability decreased by increasing the oil concentration.

Oil concentration enhancement however, increased the systems structure but resulted in larger number mean diameter that may be due to insufficient gum concentration to cover whole oil droplets retarding their movement and aggregation.

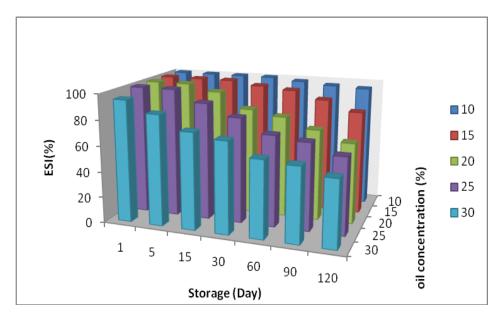


Fig. 1: Emulsion stability indexes after 120 days storage at 25 °C for emulsions containing different oil concentration

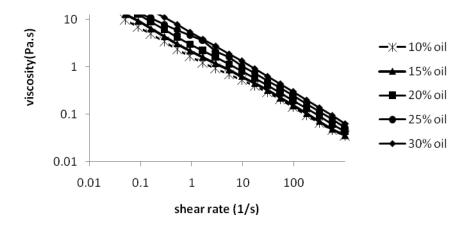


Fig. 2: Effect of oil concentration on apparent viscosity of emulsions containing different oil concentration in the presence of 0.5% w/w GT

Table 1: Power law parameters of emulsions containing different oil concentration in the presence of 0.5% w/w GT

Sample/concentration (%)	Power law parameters						
	a (pa.s ⁿ)	b	\mathbb{R}^2	s (n-1)			
10	2.01 ^e	0.42 ^a	0.99	0.1			
15	2.2^{d}	0.43^{a}	0.99	0.04			
20	3.4°	0.36^{b}	0.99	0.001			
25	4.64 ^b	0.35^{b}	0.98	0.12			
30	5.73 ^a	0.35 ^b	0.99	0.11			

Values with different letters (a–e) in each column are significantly different (p<0.05)

Table 2: Particle size analysis of emulsions containing different oil concentration in the presence of 0.5% w/w GT

Oil concentration (%)	Droplet characteristic							
	d (0.1)	d (0.5)	d (0.9)	d (4,3)	d (1,0)	span		
10	4.5°	48.94 ^b	178.31°	75.45 ^d	1.36 ^a	3.55°		
15	4.63°	45.70°	196.74 ^a	89.22 ^a	1.43 ^a	4.18^{b}		
20	5.03 ^b	48.12 ^b	185.35 ^b	78.98 ^c	1.86 ^b	3.73°		
25	4.89 ^b	33.96^{d}	168.76^{d}	81.37 ^b	2.18 ^c	4.96^{a}		
30	5.73 ^a	53.84 ^a	191.79 ^a	85.80^{a}	2.43^{d}	3.46^{c}		

Values with different letters (a–d) in each column are significantly different (p<0.05)

Conflicts of interest

All the authors of this article declare that they have no conflict of interest.

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