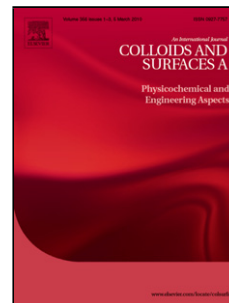


## Accepted Manuscript

Title: Formulation and optimization of emulsions based on bitter fennel essential oil and EO/BO block copolymer surfactant

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PII: S0927-7757(17)30676-3  
DOI: <http://dx.doi.org/doi:10.1016/j.colsurfa.2017.07.027>  
Reference: COLSUA 21801

To appear in: *Colloids and Surfaces A: Physicochem. Eng. Aspects*

Received date: 30-9-2016  
Revised date: 14-3-2017  
Accepted date: 8-7-2017

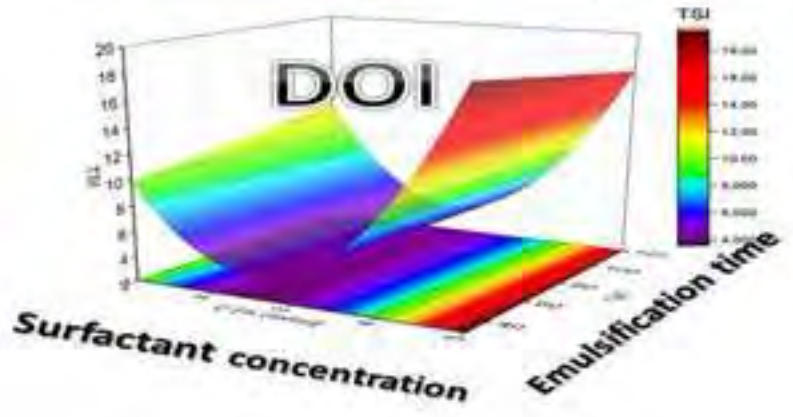
Please cite this article as: R. Llinares, P. Ramirez, J. Carmona, F. Carrillo, J. Munoz, Formulation and optimization of emulsions based on bitter fennel essential oil and EO/BO block copolymer surfactant, *Colloids and Surfaces A: Physicochemical and Engineering Aspects* (2017), <http://dx.doi.org/10.1016/j.colsurfa.2017.07.027>

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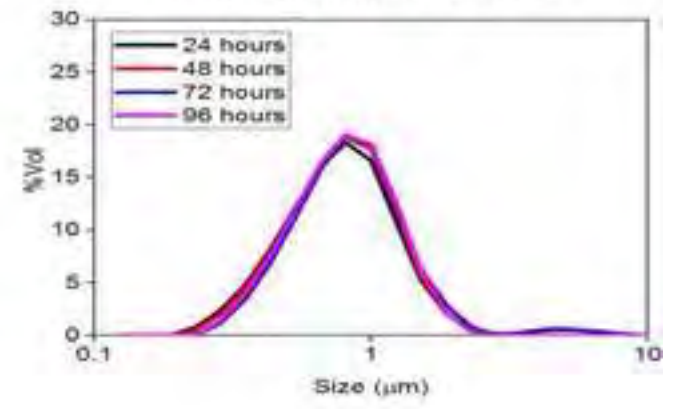
**Eco-friendly**

Bitter fennel essential oil



**Optimal Methodology**

**Stable emulsions with submicron sizes**



## Highlights

- Submicron droplet size bio-based emulsions were obtained by rotor/stator homogenizer.
- The influence of emulsification time and surfactant concentration in emulsion stability was study by surface response methodology.
- Creaming turned out to be the main destabilization mechanism.
- High pressure homogenizer or addition of rosin gum to the oil phase led to emulsions with lower droplet sizes.
- Nevertheless, higher span values are also obtained leading to more unstable emulsions.

# FORMULATION AND OPTIMIZATION OF EMULSIONS BASED ON BITTER FENNEL ESSENTIAL OIL AND EO/BO BLOCK COPOLYMER SURFACTANT

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## Abstract

The influence of EO/BO block copolymer surfactant concentration in the range (1-5 % (m/m)) and emulsification time on the emulsion stability and droplet size distribution of bitter fennel essential oil-in-water emulsions has been studied.

Essential oils are promising material to be used for emulsion formulation with applications in cosmetics, food industry and agrochemicals. In the latter field these emulsions may be used as matrices for pesticides, where essential oils play the role of an eco-friendly solvent.

A two-factor central composite design was conducted in order to optimize the emulsion formulation and processing. Emulsion stability has been studied as function of ageing time, by multiple light scattering (Turbiscan Lab-expert, Formulation) and laser diffraction (Mastersizer X, Malvern). The surface response methodology allowed us to obtain a formulation comprising minimum droplet size and maximum stability by using a single step rotor/stator homogenization process.

Creaming turned out to be the main destabilization mechanism. In order to improve emulsion stability the influence of high pressure homogenization (M110P, Microfluidics, Massachusetts, USA) and addition of a weighting agent

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(rosin gum) was studied. Although lower droplet sizes were obtained in both cases ( $D_{3,2}$  were in the range  $0.30 - 0.35 \mu m$ ), emulsions were more unstable maybe due to Ostwald ripening phenomenon.

*Keywords:* bitter fennel, essential oil, emulsion, design of experiments, rosin gum, microfluidizer

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## 1. Introduction

Recently, bio-based products are gaining more and more attention and international institutions, such as the European Commission[1] includes the development of new kinds of these products as a part of its strategy for sustainable growth. One important field in this area is the replacement of traditional organic solvents for new bio-based ones [2, 3]. According to the European Committee for Standardization bio-based products are defined as "products that are wholly or partly derived from materials of biological origin, excluding materials embedded in geological formations and/or fossilised" [4].

10

Essential oils are hydrophobic compounds of biological origin that possess antibacterial, antifungal, antiviral and antioxidant activity. Their hydrophobicity hinders many of their potential applications[5], such as preservative or flavouring agents in food, cosmetics or medicines[6]. Furthermore, volatility of essential oils is a major drawback for their applications. One way to diminish this problem is by producing stable emulsion formulations that will reduce evaporation[7].

15

In the present work, emulsions containing essential bitter fennel oil (*F. vulgare* Mill. var. *vulgare*) as bio-based solvent have been prepared. The composition of essential bitter fennel oil is related to the origin, the harvesting period, the processing conditions and the part of the plant it is extracted from [8–10]. Main components found are anethole, fenchone, estragole limonene and/or  $\alpha$ -Phellandrene. They are highly hydrophobic and in the case of anethol and

20

25 limonene it has been reported that their O/W emulsions are destabilized by  
Ostwald ripening[11, 12]. Nevertheless, it has been recently reported that very  
stable emulsions are obtained when the dispersed phase consists of a mixture  
of two solvents such as N,N-dimethyl decanamide and  $\alpha$ -pinene or d-limonene  
in a ratio of 75/25[13, 14]. Hence, the presence of a mixture of two or more  
30 solvents in the dispersed phase, (as in the case of fennel oil) is able to produce  
synergistic effects such as the reduction of emulsion destabilization by Ostwald  
ripening or the decrease of the interfacial tension between the two phases.

Furthermore, in order to obtain a “green emulsion” the surfactant has to be  
35 carefully chosen. In this study, an EO/BO block copolymer (DOWFAX 20B102)  
has been used. According to its manufacturer this surfactant is non-volatile, wa-  
ter soluble and readily biodegradable. This kind of block copolymers are widely  
used in agrochemical formulations[15].

40 Obtaining small droplet sizes and narrow distributions is a major goal in the  
emulsification process. This objective is usually achieved by mechanical energy  
input to the system with different devices such as rotor-stator or high-pressure  
homogenizers[16]. The former produces turbulent and elongational flows be-  
tween the rotor and the stator whereas the later use high-pressure pumps to re-  
45 duce the droplet sizes of coarse emulsions previously prepared with rotor-stator  
devices, by means of shear, cavitation, elongational, and turbulent flows[17-  
19].

Emulsions are thermodynamically unstable colloidal dispersions, which may  
50 undergo one or several destabilization mechanisms such as flocculation (aggre-  
gation of droplet without rupture of the interface), coalescence (merging of oil  
drops with the rupture of interface), creaming/sedimentation (gravitational sep-  
aration induced by density difference of the phases) and Ostwald ripening (the  
solubility of the material contained in the smaller droplets is higher than larger  
55 ones, thus the material diffuses through the continuous phase from the smaller

to the bigger droplets due to this concentration gradient)

In the present work a design of experiment strategy has been used to determine the optimum surfactant concentration and emulsification time in order to obtain submicron stable fennel oil-in-water emulsions by means of simple rotor-stator homogenization. Furthermore, high pressure homogenization was also used with the most stable emulsions in order to reduce the size of droplets, which would likely enhance the physical stability of the emulsions.

## 2. Materials and methods

### 2.1. Materials

Bitter fennel essential oil ( $897 \text{ g/L}^{-1}$ ) was kindly provided from Bordas Chinchurreta S.A. and was used as the dispersed phase in the O/W emulsions. An aqueous solution of an EO/BO block copolymer surfactant (DOW FAX 20B102,  $M_w = 800 \text{ g/L}^{-1}$ , HLB = 11-13) was used as continuous phase. The copolymer surfactant was kindly provided by Dow Chemical. Rosin gum (Sigma-Aldrich®) is an exudate gum which is solid at room temperature. It is a mixture of different acids such as abiatic acid, palaustic acid, neoabetic acid, being the abiatic acid the most abundant. Milli-Q water was used to prepare the continuous phase.

### 2.2. Chromatographic analysis

Samples for GC-MS analysis were prepared by dissolving  $100 \mu\text{L}$  of essential oil in  $1 \text{ mL}$  of dichloromethane. The chromatographic analysis of bitter fennel oil was carried out following the procedure of Miraldi[9] on a Thermo scientific TSQ 8000 triple quadrupole gas chromatography–mass spectrometer (GC–MS/MS) equipped with a Supelcowax-10 column ( $30 \text{ m } 0.32 \text{ mm } 0.25 \text{ m}$ ) and helium as carrier gas. The composition is given in table 1. The major components found are in agreement with those found in previous studies[10], although the anethole concentration is lower than usually reported[8, 9].

Compound	relative concentration (%)
Limonene	23.0
Anethole	22.0
$\alpha$ - Phellandrene	15.0
Fenchone	12.9
$\alpha$ - pinene	5.7
Estragole	4.8
$\beta$ - myrcene	3.4
$\beta$ - pinene	2.7
$\gamma$ -terpinene	2.3
$\beta$ - Phellandrene	2.2

Table 1: Percentage composition of the bitter fennel essential oil used in the present study

### 2.3. Droplet size distribution measurements

85 Droplet size distribution measurements were carried out by using laser diffraction technique (Mastersizer X, Malvern, Worcestershire, United Kingdom). Sauter diameter was used to compare the mean droplet diameter of different emulsions, and it is given by:

$$D_{3,2} = \frac{\sum_{i=1}^N n_i d_i^3}{\sum_{i=1}^N n_i d_i^2} \quad (1)$$

90 where  $d_i$  is the droplet diameter,  $N$  is the total number of droplets and  $n_i$  is the number of droplets having a diameter  $d_i$ .

Furthermore, span values were used to study the distribution width of droplet sizes. Span is defined by the following equation:

$$\text{Span} = \frac{D(\nu, 0.9) - D(\nu, 0.1)}{D(\nu, 0.5)} \quad (2)$$

95 where  $D(\nu, 0.9)$ ,  $D(\nu, 0.5)$  and  $D(\nu, 0.1)$  are the highest droplet size containing the 90%, 50% and 10% of volume of dispersed phase, respectively.



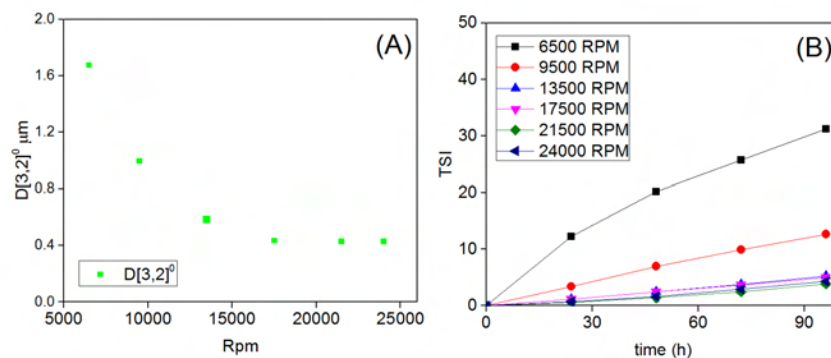


Figure 1: A) Sauter mean diameter as a function of homogenization rate. B) Turbiscan Stability Index as a function of homogenization rate. Bitter fennel essential oil-in-water emulsion with  $C = 3\%$  (m/m) and  $\phi = 10\%$ . Symbols are the mean of three replicates.

#### 2.4. Multiple light scattering

The physical stability of the emulsions were evaluated by means of multiple light scattering measurements (Turbiscan Lab Expert). Since, emulsion can undergo different destabilization processes, the global TSI (turbiscan stability index) parameter has been used to compare emulsion stability. This parameter has been used in previous works[12, 14] and is defined by the following equation[20]:

$$TSI = \sum_j |scan_{ref}(h_j) - scan_i(h_j)| \quad (3)$$

where  $scan_{ref}$  and  $scan_i$  are the initial backscattering and the backscattering value after a certain time, respectively.  $h_j$  are the different heights of the measurements cell at which backscattering has been measured and, hence,  $TSI$  is the sum of all the scan differences at a given time.

#### 2.5. Emulsification method

Diluted oil-in-water emulsions (10 wt% dispersed phase) were prepared with a rotor-stator homogenizer (Ultra turrax T-25/KV 11, IKA Instruments, Germany). The oil phase was mixed with previously thermostated continuous

phase. After emulsification the emulsion temperature was 5-10 °C higher than the initial temperature of the continuous phase. In previous tests it was observed that droplet size decreased when decreasing the initial temperature of the continuous phase from 25°C to 5°C. This is likely due to a lesser recoalescence phenomenon when decreasing emulsification temperature [21, 22]. Hence, a initial temperature of the continuous phase of 5°C was used in this study.

Several emulsion were produced varying the homogenization rate at a constant emulsification time of 60 seconds and for a surfactant concentration of 3%wt. Figure 1A illustrates that the Sauter's mean diameter dropped with homogenization rate but levelled off above 17500 rpm. Interestingly, figure 1B shows that the lower TSI values were achieved at homogenization rates in the 17500 - 24000 rpm range, congruently with the lower initial Sauter mean diameters obtained. An intermediate homogenization rate well within the optimum range (21500 rpm) was chosen to study the effect of homogenization time on droplet size distribution and physical stability of emulsions.

Finally, Table 2 summarizes the processing conditions used in the present study.

Formulation	
Surfactant concentration	1-5 wt%
Dispersed phase	10 wt%
Emulsification Process	
Temperature	5°C
Homogenization rate	21500 r.p.m.
Emulsification time	30-120 s

Table 2: Emulsification conditions

The most stable emulsion obtained by rotor/stator homogenization was further passed through an air-driven microfluidizer (Model M-110P, Microfluidics,

USA) operating at 7500 psi (517.1 bar) with a F12Y interaction chamber. The interaction chamber was at room temperature.

### 2.6. Design of experiments

135 In order to obtain an optimal formulation of bitter fennel oil-in-water emulsions and to determinate the possible existence of synergistic effects between the analysed factors, a central composite design with four replications of the centre point has been used. In this work surfactant concentration ( $C$ ) and emulsification time ( $t$ ) have been chosen as process factors. The quadratic model for the  
140 response variables is given by:

$$Y = \beta_0 + \beta_1 C + \beta_2 t + \beta_3 C^2 + \beta_4 t^2 + \beta_5 Ct \quad (4)$$

Each factor was measured at five different levels (-1.414, -1, 0, 1, 1.414). Table 3 summarizes the experimental design.

As response variables Sauter mean diameter ( $D_{3,2}$ ) and Turbiscan Stability Index ( $TSI$ ) were used. Experiments were randomly carried out in order  
145 to minimize the effects of order related errors in the observed responses. The centre point in the design was repeated four times to calculate the repeatability of the method [23] and to check the fitting quality of the mathematical model [24]. For model construction, terms with  $p > 0.05$  were removed and the analysis was recalculated without these terms. The suitability of the models was  
150 determined by using coefficient of determination (adjusted  $R^2$ ) and the lack of fit test (Fisher-Snedecor coefficient,  $F_{lof}$ ). Joklegar and May[25] proposed that  $R^2$  should be higher than 0.80 to obtain a good fitting.

## 3. Results and discussion

### 155 3.1. Droplet size distribution

Central composite design can be used as part of response surface methodology which aim is to use designed experiments to obtain an optimal response.

Sample code	Surfactant conc. (%wt), $C$	Emulsification time (s), $t$
E1	1.59	43
E2	4.41	43
E3	1.59	107
E4	4.41	107
E5	5	75
E6	1	75
E7	3	30
E8	3	120
E9	3	75
E10	3	75
E11	3	75
E12	3	75
E13	3	75

Table 3: Experimental design with five central points

The Sauter mean diameter was not significantly influenced by emulsification time, hence the quadratic model equation became only a function of surfactant concentration according to:

$$D_{3,2} = 0.48 - 0.27C + 0.17C^2 \quad (5)$$

where  $C$  was in the range (-1.414, 1.414).

The model employed was shown to be adequate with no significant lack of fit ( $F_{crit} > F_{lof}$  with  $\alpha = 0.05$ ) and a value of adjusted  $R^2$  of 0.984.

Several emulsions were prepared at intermediate surfactant concentrations in the range 1-5 % (m/m) and at the lowest emulsification time tested since the latter variable did not significantly influence Sauter mean diameters. Figure 2

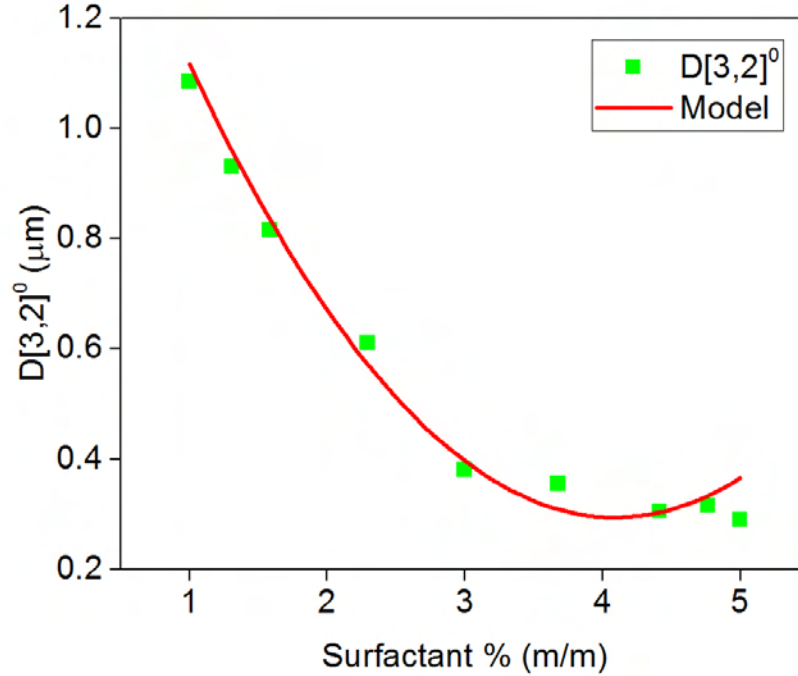


Figure 2: Sauter mean diameter as a function of surfactant concentration for bitter fennel essential oil-in-water emulsion with 30 s of emulsification time and  $\phi = 10\%$  (m/m). The line shows data fitting to the model (Eq. 4). Symbols are the mean of three replicates.

shows the Sauter diameter of these emulsions as a function of surfactant concentration. It is shown that the model equation fits reasonably well all the experimental values. Sauter mean diameters decreased with surfactant concentration in the (1-4) % (m/m) concentration range but levelled off above 4 % (m/m). The interfacial adsorbed amount the solubility would decrease at high temperature due to dehydration on account of rupture of hydrogen bonds, similarly to the reason behind the appearance of the so-called cloud point of surfactant can be approximately calculated for polydisperse emulsions from equation 6 [21]:

$$\Gamma_m = \frac{CD_{3,2}}{6\phi} \quad (6)$$

Then, given  $\phi$ ,  $C$  and the experimental value of the Sauter diameter ( $D_{3,2}$ ),  $\Gamma_m$

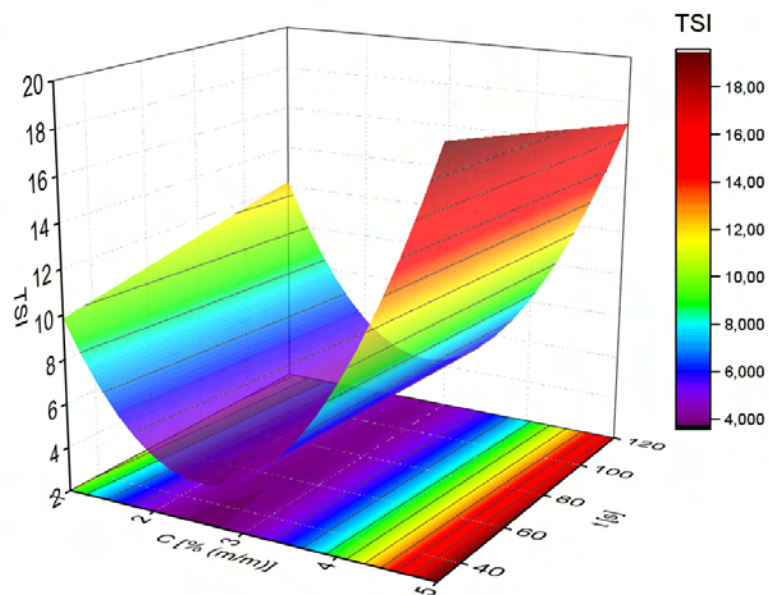


Figure 3: Response surface 3D plot and model equation of Turbiscan Stability Index as a function of surfactant concentration and emulsification time for bitter fennel essential oil-in-water emulsion with  $C=2.65\%$  (m/m) and  $\phi=10\%$  (m/m).

is readily calculated from the above equation. Table shows  $\Gamma_m$  as a function of  $C$ . All calculated  $\Gamma_m$  values are higher than those reported for these kind of polymeric surfactants[26] ( $\Gamma_m$  usually falls between 3 to  $8 \text{ mg} \cdot \text{m}^{-2}$ ). Nevertheless, since for the concentration range 1-4% (m/m) droplet size is mainly affected by surfactant concentration it is likely that insufficient surfactant has been added, and surfactant multilayers are probably formed, hence  $\Gamma > \Gamma_m$ . For surfactant concentration above 4% (m/m) the droplet size is relatively independent of surfactant concentration indicating that an excess of surfactant is added and, therefore droplet size will be strongly dependent on the emulsification process.

Therefore a minimum droplet size value was obtained for concentrations higher than 4% m/m.

$C / \% \text{ (m/m)}$	$D_{3,2} / \mu\text{m}$	$\Gamma_m / \text{mg} \cdot \text{m}^{-2}$
1.0	1.12	17.4
1.4	0.92	19.1
1.6	0.83	19.4
2.2	0.65	20.3
3.0	0.40	18.2
3.6	0.35	19.1
4.4	0.30	20.0
4.7	0.31	22.1
5.0	0.29	22.0

Table 4: Maximum surface coverage according to equation 6

### 190 3.2. Emulsion stability

Considering that emulsion destabilization is usually provoked by the cooperative effect of different mechanisms, such as flocculation, creaming, coalescence or Ostwald ripening, the *TSI* was chosen to assess the physical stability of the emulsions studied. This parameter has the advantage that it quantifies the overall emulsion destabilization as a function of ageing time. However, it does not give information about the predominance of one mechanism over another. In the present study the emulsion stability was monitored by following the evolution of the *TSI* for four days.

200 From central composite design analysis it was observed that *TSI* is not influenced by the linear or quadratic term of emulsification time. However, the interaction term has to be taken into account. Hence, the model equation is given by:

$$TSI = 4.3 + 2.5C + 5.1C^2 - 0.6Ct \quad (7)$$

where  $C$  and  $t$  were in the range (-1.414, 1.414).

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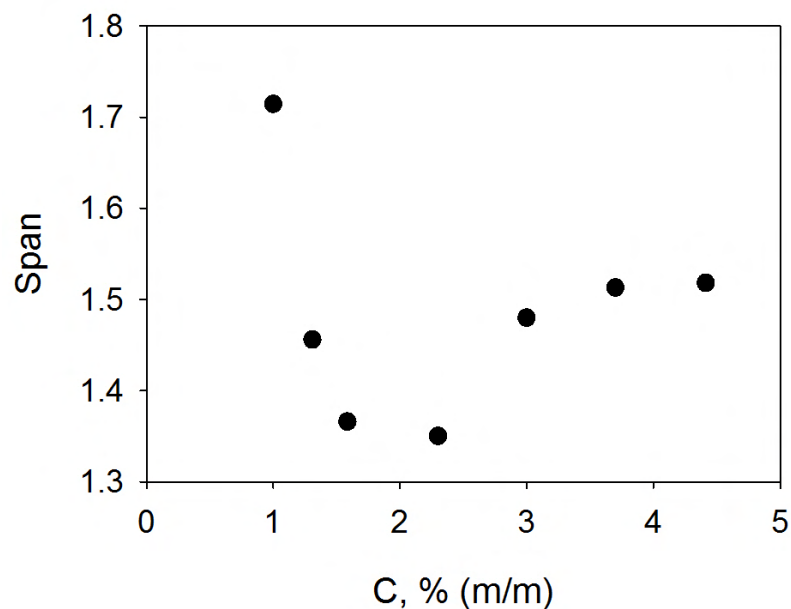


Figure 4: Span as a function of surfactant concentration for bitter fennel essential oil-in-water emulsion with 30 s of emulsification time and  $\phi = 10\%$  (m/m). Symbols are the mean of three replicates.

The model employed was shown to be adequate with no significant lack of fit ( $F_{crit} > F_{lof}$  with  $\alpha = 0.05$ ) and a value of adjusted  $R^2$  of 0.981.

The minimum value of  $TSI$  was predicted by the mathematical model at a surfactant concentration of 2.6 % (m/m) and an emulsification time of 33 s. It is worth noting that the most stable emulsion (the one with the lowest  $TSI$  value) was obtained for a surfactant concentration of 2.6% (m/m). This concentration value is lower than the limiting surfactant concentration (4.0% (m/m)) needed to achieve the lowest Sauter mean diameter. This could be related to an increase in polydispersity for more concentrated emulsion, which in turn yielded an increase in the creaming rate due to higher values of the effective packing



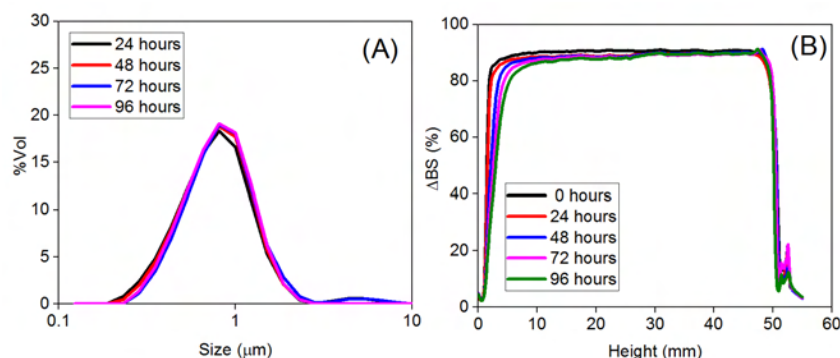


Figure 5: A) Droplet size distributions of emulsions prepared with a rotor-stator homogenizer as a function of ageing time. B) backscattering versus container height as a function of time. Bitter fennel essential oil-in-water emulsion with  $C=2.65\%$  (m/m) and  $\phi=10\%$  (m/m).

parameter [21, 27]. Figure 4 shows the span (a measurement of droplet size polydispersity) for the emulsions of figure 2 exhibiting a minimum value at a concentration close to that predicted by *TSI* data. This seems to indicate that the *TSI* parameter is clearly influenced by span values, i.e. by the droplet size polydispersity.

### 3.3. Optimum formulation

Bitter fennel oil emulsions with submicron Sauter mean diameters and enhanced physical stability were prepared and analysed Figure 5A illustrates that the droplet size distribution remained almost unaffected for 4 days ageing time. Although, *TSI* values used to study emulsion stability do not differentiate between the different destabilization processes a close look at the multiple light scattering data allow us to identify creaming as the main destabilization process due to the decrease of backscattering values at the lowest part of the vial.

This gravitational separation process can be quantified by measuring the creaming rate ( $v$ ) from the slope of  $H_s$  vs. time, where  $H_s$  is the height of the serum layer [28, 29] from the data of Figure 5B a value of  $v = 0.025 \text{ mm} \cdot \text{h}^{-1}$

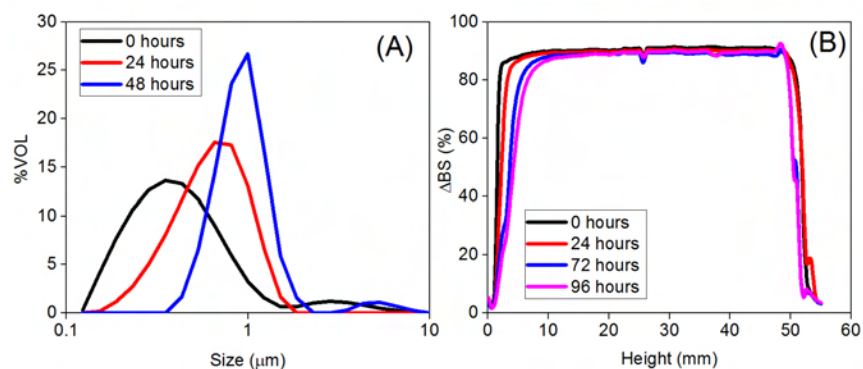


Figure 6: A) Droplet size distributions of emulsions prepared with Microfluidizer M110P at 7500 psi and one pass as a function of ageing time B) backscattering versus container height as a function of time. Bitter fennel essential oil-in-water emulsion with  $C=2.65\%$  (m/m) and  $\phi=10\%$  (m/m).

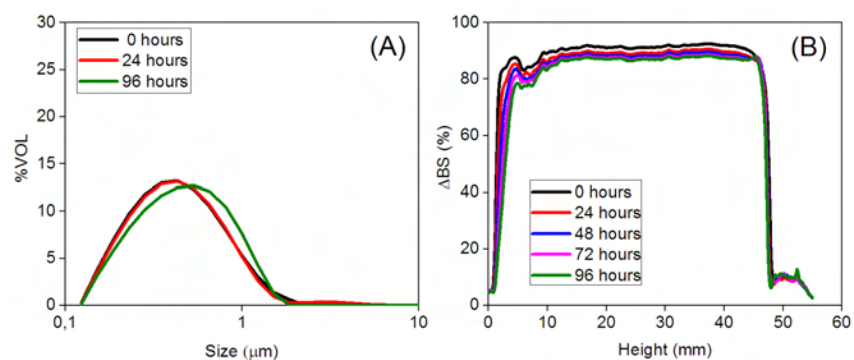


Figure 7: A) Droplet size distributions of emulsions prepared with a rotor-stator homogenizer as a function of ageing time B) backscattering versus container height as a function of time. Bitter fennel essential oil-in-water emulsion with  $C=2.65\%$  (m/m) and  $\phi=10\%$  (m/m) with a oil phase composition of 30% (m/m) rosin gum and 70% (m/m) bitter fennel essential oil.

235 was calculated

As a rule creaming rate of emulsions is usually expressed as a function of the creaming rate derived from Stokes' law multiplied by a factor that is function

of the volume fraction:[29, 30]

$$v = v_{Stokes} \cdot K(\phi) \quad (8)$$

240 and, for dilute emulsion the following equation can be used to calculate the creaming rate:[29]

$$v = v_{Stokes}(1 - 6.55\phi) \quad (9)$$

where  $v_{Stokes}$  is the creaming rate from Stokes' law:

$$v_{Stokes} = \frac{gd^2(\rho_2 - \rho_1)}{18\eta_1} \quad (10)$$

g is the acceleration due to gravity, d is the diameter of the particle,  $\rho_1$  and  $\rho_2$  are the densities of dispersed and continuous phases, respectively and  $\eta_1$  is the viscosity of the continuous phase. Considering  $\rho_1 = 1000kg \cdot m^{-3}$ ,  $\rho_2 = 897kg \cdot m^{-3}$  245 and  $\eta_1 = 1 \cdot 10^{-3}Pa \cdot s$  a value of  $v = 0.023 mm \cdot h^{-1}$  was obtained in good agreement with the experimental value obtained which clearly indicates that creaming is the main destabilization process.

In order to inhibit this unwanted phenomenon several strategies can be 250 adopted, such as to diminish droplet size by further processing with more powerful devices (high pressure homogenization, ultrasounds, etc.); reducing the driving force for gravitational separation, that is density difference by adding a weighting agent; or modifying the viscosity of the continuous phase. The former two strategies were explored in this study and main results are presented below:

### 255 3.3.1. High pressure homogenization

A high-pressure homogenizer with microfluidizer technology, model M110P was used in order to reduce the droplet size of the coarse rotor-stator emulsion(Dickinson, 1988). The microfluidizer operated at 7500 psi and 20°C and coarse emulsions were passed from 1 to 3 times through the homogenizer.

260

Figure 6 shows the evolution of droplet size distribution for 48 hours for 1 pass-emulsion (emulsions with 2 or 3 passes show the same behaviour and are not shown for the sake of clarity). Although, lower Sauter diameter were obtained

( $D_{3,2}$  dropped from 0.58 to 0.30  $\mu m$ ) droplet size distribution shifted toward  
265 higher droplet sizes showing a faster destabilization compared to previously  
obtained with simple rotor-stator homogenizer. This result can be explained by  
the increase in polydispersity (span value increase from 1.17 for rotor-stator to  
1.525 for microfluidics). Moreover, an initial bimodal distribution was observed  
with two main distributions one of large droplets higher than 2  $\mu m$  and another  
270 one with sizes well below 1  $\mu m$  which will trigger the occurrence of Ostwald  
ripening destabilization.

### 3.3.2. Addition of a weighting agent

It has been shown that addition of a weighting agent can reduce the creaming  
rate[31]. In recent studies rosin gum was used as second component of dispersed  
275 phase in d-limonene-in-water emulsions given rise to very stable emulsions[27].  
Hence, the optimum emulsion was modified by adding rosin gum (30% m/m)  
as weighting agent to the dispersed phase.

A decrease in the emulsion droplet size is observed in the droplet size distri-  
280 bution shown in figure 7 (from 0.58 to 0.35  $\mu m$  when rosin gum is added). This  
decrease in droplet size is likely to be due to the fact that rosin gum adsorbed  
at the interface as has been recently reported for d-limonene-water interface.  
Hence, rosin gum acts as a co-surfactant reducing interfacial tension and faci-  
litating the emulsification process. Nevertheless, emulsion stability is lower than  
285 emulsions prepared without rosin gum. Moreover, the decrease of backscatter-  
ing signal with aging time throughout the vial height point to a droplet size  
increase which is likely due to Ostwald ripening.

## 4. Conclusions

Bitter fennel essential oil-in-water emulsions of submicrom range were ob-  
290 tained by rotor/stator homogenizer by using a diblock EO/BO copolymer as  
emulsifier. The droplet size distribution and stability of these emulsions have  
been analysed by using a design of experiment strategy by varying surfactant

concentration and emulsification time. According to the statistical analysis of the data, emulsification time has little influence in droplet size distribution and emulsion stability. The optimum surfactant concentration to obtain the lowest Sauter mean droplet diameter and the longest physical stability as a function of ageing time was estimated to be ( $C = 2.65\%$  (m/m)) by the mathematical model used. Nevertheless, this formulation involved the onset of a creaming destabilization process as demonstrated and monitored by multiple light scattering tests. In order to reduce the creaming rate two strategies have been explored: (i) reducing the droplet size distribution by using a high pressure homogenizer and (ii) addition of a weighting agent (rosin gum) to the dispersed phase. Both strategies led to lower droplet size values but with higher span values which increase the creaming destabilization of the emulsions. Furthermore, the interaction between rosin gum and bitter fennel essential oil did not prevent the occurrence of destabilization by Ostwald ripening. Further investigations based on the choice of alternative weighting agents or viscosity modifiers of the dispersed phase are required to improve the physical stability of the emulsions studied.

### 310 **Acknowledgements**

The financial support received (Project CTQ2015-70700-P) from the Spanish Ministerio de Economía y Competitividad and from the European Commission (FEDER Programme) is kindly acknowledged. The authors are also grateful to Bordas Chinchurreta and KAO for providing materials for this research.

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