Production of more sustainable emulsions formulated with eco-friendly materials

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Abstract

Sustainable development involves the search for new products with a low environmental impact. Hence, the aim of this work is to obtain stable and concentrated aqueous emulsions containing bitter fennel oil and a biomass-derived emulsifier by studying different processing variables and techniques. Firstly, the effects of the application of a premix step previous to homogenization and of the geometry of the high-energy rotor-stator device used (Silverson L5M or Ultraturrax T50) on the droplet size distribution (DSD) and physical stability (PS) of emulsions were investigated. The use of a premix worsens both the physical stability and the average droplet diameters of the emulsions, the most stable emulsion being that obtained with the Silverson L5M alone. Secondly, the stability of this emulsion was improved. To achieve this goal, this coarse emulsion was microfluidized at different pressures (from 5000 to 25000 psi), reaching submicron sizes and monomodal distributions, except for that subjected to 25000 psi which resulted in clear overprocessing. Creaming and oiling off were the main destabilization processes. The emulsion exhibiting the lowest droplet sizes and the best physical stability was prepared at 20000 psi. This work contributes to the development of sustainable agrochemical prototypes.

Keywords: Bitter fennel oil, Wheat derived surfactant, Emulsion, Premix in primary homogenization, Microfluidizer, Physical stability.

Introduction

In order to reduce both human health and environmental risks, scientists have a great effort to work out future strategies with respect to green chemistry (Durand et al., 2016). One of them is the incorporation of eco-friendly materials in the design of new products. For this reason, sustainability has become an important requirement for solvents, leading to the need to develop new green solvents. The non-polar characteristic of some of these organic solvents means that they cannot be dispersed directly in another aqueous phase (Israelachvili, 2011). Hydrophobic compounds could be incorporated as functional or active ingredients in a wide variety of colloidal systems with application in sectors such as food or agrochemicals. An important application of this type of system in the field of agrochemicals is the use of emulsions of essential oils, which can be used as a matrix for pesticides, where the essential oil functions as a solvent friendly to the environment. In previous works, emulsions containing essential oils such as alpha-pinene (García et al., 2014; García et al., 2015), limonene (Trujillo-Cayado et al., 2016) or thyme essensial oil (Martin et al., 2018) were studied for this purpose.

Another important issue is that the fact of considering the use of the biodegradable raw materials leads to obtaining both a final product with little impact on the environment and waste which is less harmful to it (Bom et al., 2019). As indicated Saéz-Martinez et al (2016) the future must be made up of green, sustainable and recyclable chemicals. In the present work, in order to achieve this purpose, emulsions containing bitter fennel essential oil as an ecological solvent were produced. The bitter fennel plant is a common perennial hemicryptophyte from the Mediterranean basin whose main components are estragole

and trans-anethole in addition to the cyclic monoterpenes of fenchone and limonene (Gross et al, 2002).

Frequently, essential oils have been utilized as natural preservatives and as fragrances in cosmetic products, but as a result of their antimicrobial and antioxidant properties, new applications are emerging for them in sectors such as food or agriculture (Rodríguez-Rojo et al., 2012). Their hydrophobicity, as mentioned above, makes many of their applications difficult, although it is their volatility that supposes a greater barrier (Llinares et al., 2018). One way to reduce this problem is the formulation of stable emulsions that reduce evaporation (Rodríguez-Rojo et al., 2012). Oil-in-water emulsions are one of the main components of many commercial products. Currently, this type of emulsion can be found in foods, vitamin supplements, drugs, cosmetics, personal care items and agrochemicals. However, emulsions are, from a thermodynamic point of view, extremely unstable and the two phases that compose them tend to separate (Borwankar et al., 1992). For this reason, other components such as surfactants, proteins or thickeners are commonly used to improve emulsion stability, with the use of the surfactant playing an important role in this. The surfactant molecules tend to position themselves at the interface produced between the dispersed oil droplets and the aqueous continuous phase (Dickinson et al., 1989). Thus, several processes involved in emulsion destabilization are controlled by the presence of the surfactant, which influences electrostatic and steric repulsion and consequently, the emulsion stability (McClements, 2007). For this purpose, Applyclean 6548 has been employed as emulsifier. It is a non-ionic surfactant derived from wheat which belongs to the family of alkyl poly pentosides (APP). This surfactant has all the necessary characteristics to be considered as an ecological surfactant, since it derives from a renewable source and its production is carried out by means of a process that respects the environment (Trujillo-Cayado et al., 2018). In fact, this surfactant, developed by the

French company Wheatoleo, has ECOCERT certification. Its main characteristics include its low toxicity, its rapid biodegradation and its HLB, which is between 9.0 and 9.5. In addition, it should be noted that the use of this surfactant, which is obtained from wheat straw, avoids the waste accumulation in the environment and converts these wastes into added-value materials. Furthermore, as Lin et al (2013) point out, its raw material does not compete with food production.

Another point to take into account to increase of these emulsions sustainability is the use of the most friendly production strategy. This is to say to perform a cleaner production in which the environmental contamination is reduced as Krolczyk et al. purposed in a previous work (Krolczyk et al., 2017). Recent works have carried out this aim centering their studies in ecological procedures (Mia, et al., 2018). It should be noted that today it is so important to provide solutions for environmental degradation that there are numerous studies that are being developed related to sustainable production. Among the most recent ones, the one carried out by Correa et al. (2019) for the biofuel production, the study of Nidheesh and Kumar (2019) for the cement and steel production or that of Mark et al. (2019) for the production of phenolic compounds. In this way, Bom et al., (2019) in their review about sustainability in the cosmetic industry considered several processing variables such as the use of the same equipment in process in which several steps are involved, the production energy optimization, the reduction of the washing water, etc. The same points can be used in other industries. In addition, it is worth making an effort to find out the devices which produce less impact on the environment. As Krolczyk et al (2019), reported, this fact can be the opener for a cleaner and friendly production. For this purpose, the main objective of this work is to evaluate different processing conditions to produce an environmentally friendly emulsion from a mixture whose main components are bitter fennel essential oil and water, in order to select the most sustainable processing protocol that provides greater emulsion stability. Firstly, the influence of a premix stage in the processing of oil-of-fennel-in-water emulsions was studied. For this, four experiments were carried out. On the one hand, two systems were subjected to a premixing process with a low-energy rotor-stator equipment prior to the emulsification with high-energy rotor-stator devices (two different geometries) and on the other hand, two systems in which the emulsification was carried out directly, without

any type of premix. The importance of this stage of the study lies in determining the number of steps of the most sustainable processing. Subsequently, the stability of the emulsions obtained as a function of aging time was studied by means of laser diffraction and multiple light scattering techniques. The most stable emulsion produced during this stage was then improved with the microfluidization equipment (Microfluidizer M-110P). The influence of pressure on its properties was analyzed. In this way, the optimal processing conditions were established for the emulsification of a mixture whose main elements are bitter fennel essential oil, wheat biomass-derived surfactant and water which could be used as a prototype for sustainable agrochemicals. An analysis of both processing variables led to the optimization of the processing protocol in order to achieve the most respectful with the environment.

Materials and methods

Materials

Bitter fennel essential oil (density: 0.897 Kg/m^3) kindly supplied by Destilaciones Bordas Chinchurreta S.A was used as dispersed phase. Applyclean 6548 (D-xylofuranose, oligomeric tetradecyl and octodecyl glycoside, C14, C18 alcohol) (Martín et al., 2018), produced by Wheatoleo, was utilized as emulsifier. In order to preserve the emulsions, sodium azide (Panreac) was included. Milli- Q water was used to complete the formulation.

Studied emulsions contained 40 wt% oil phase, 4 wt% surfactant, 0.1 wt% sodium azide and Milli- Q water as continuous phase.

Preparation of emulsions

The continuous phase was prepared by mixing the necessary amount of sodium azide in Milli-Q water at room temperature by means of a magnetic stirring plate (SM 162, Stuart, Scientific Laboratory Supplies). Subsequently, the dispersed phase was obtained by

dissolving the appropriate quantity of Applyclean 6548 in bitter fennel essential oil at 70ºC for 15 minutes. For this purpose, a Phoenix (Thermo-Scientific) bath was employed.

Once both dispersed and continuous phases were prepared, four different emulsions were obtained using the following protocols:

1. IKA-UT50 emulsion. These emulsions were produced by adding the dispersed phase to the continuous phase at 400 rpm for one minute using an IKA-VISC MR-D1 low-energy rotor-stator homogenizer (IKA Labortechnik, Germany) and then continuing for one additional minute at the same rate. This emulsion was immediately homogenized with a high speed rotor-stator homogenizer (Ultraturrax T50) at 2000 rpm for one minute and then 30 s at 6000 rpm.
2. IKA-SL5M emulsion. The process used in this case was similar to that of the previous emulsion. In this protocol, a Silverson L5M rotor-stator homogenizer was utilized instead of the Ultraturrax T50.
3. UT50 emulsion. The dispersed phase was added to the aqueous phase for 1 minute at 2000rpm by means of the Ultraturrax T50 homogenizer. Lastly, 6000 rpm for 30 s was applied as a final homogenization.
4. SL5M emulsion. The processing protocol was similar to the UT50 emulsion, but the equipment used was Silverson L5M.

In a further stage, the most stable emulsion produced during the first stage was submitted to a second homogenization process by means of Microfluidizer M-110P (Microfluidics, EEUU). In this equipment, batches of 200 g were processed at different pressures (5000 psi, 10000 psi, 15000 psi, 20000 psi and 25000 psi).

At least two samples were prepared with every protocol.

Emulsion characterization

Laser diffraction

This technique was employed to determine the droplet size distribution. For this purpose, a Mastersizer 2000 (Malvern, Uk) was used. Milli-Q water was utilized as the dispersant medium. The refraction index was 1.54 for the oil dispersed phase and the refraction and adsorption indexes for the aqueous medium were 0.5 and 1.33, respectively.

In order to analyse the influence of the processing protocols on the mean droplet diameters, Sauter diameter (D[3,2]) and volume mean diameter (D[4,3]) have been employed. These parameters are defined as following:

$$D[3,2] = \sum_{i=1}^{N} n_i d_i^{\,3} \Big/ \sum_{i=1}^{N} n_i d_i^{\,2}$$

$$D[4,3] = \sum_{i=1}^{N} n_i d_i^{\,4} \Big/ \sum_{i=1}^{N} n_i d_i^{\,3}$$

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

To find out the distribution width of droplet sizes distribution, the "span" was used, which was determined as follows:

$$Span = (D[v,0.9] - D[v,0.1])/D[v,0.5]$$

where D[v, 0.9] and D[v, 0.1] stand for the 90^{th} and 10^{th} percentiles and D[v, 0.5] for the median.

In order to study the emulsion stability, these measurements were performed at 1 day, 5 days and 14 days of aging time.

At least two replicates of each test were performed at room temperature.

Multiple light scattering

Turbiscan Lab (Formulation, France) was used to evaluate the physical stability of the emulsions obtained. Tests were carried out at room temperature. Turbiscan determines backscattering and transmission as a function of the length of the cell containing the sample, but in this work only backscattering is presented as a consequence of the fact that the transmission values recorded were null.

Results and discussion

Influence of the premix and the high-energy rotor-stator device used on the microstructure and physical stability of emulsions

Figure 1 shows the influence of the processing protocols on the droplet size distribution of the emulsions containing bitter fennel essential oil as dispersed phase after 24 hours of aging. Monomodal distributions with a peak at around 2.5 μm, were exhibited for all the emulsions produced. As can be observed, emulsions obtained with a previous homogenization step presented a droplet size distribution centred on higher droplet sizes. This could be a result of a re-coalescence mechanism related to an excess of applied energy which could have provoked the breaking of several droplets. This fact led to the displacement of the curve towards higher diameters and, therefore, to emulsions with larger droplets sizes (Jafari et al., 2008; García et al. 2016). In addition, the emulsions obtained with the Silverson L5M equipment showed droplet size distributions displaced towards smaller diameters than those prepared by means of Ultraturrax T50. This result had already been observed in a previous work (Trujillo-Cayado et al., 2018). Although, under the same operation conditions, the Ultraturrax T50 device applies higher energy than Silverson L5M, the latter is more effective, probably due to the geometry of the rotor used. This finding was supported by the mean diameter values and the standard deviations exhibited in Table 1. Therefore, the best results were presented by emulsions obtained

only using Silverson 5M. From a sustainable production point of view this result is very important since involves an energy reduction and higher production efficiency (Alayón et al., 2017)

By way of example, Figure 2 illustrates the evolution of droplet size distribution with aging time for the emulsion developed by IKA-SL5M processing. As can be observed, there was no significant changes in the mean droplet sizes and, therefore, there was no destabilization by coalescence or Ostwald ripening during the evaluation time. The same result was obtained for the other emulsions investigated. This fact revealed that Appyclean 6548 perfectly fulfills its role as an emulsifier since not only made it possible to obtain concentrated emulsions, but that it was also able to satisfactorily cover the oil/water interface protecting it against the rupture.

The emulsion stability was assessed by multiple light scattering monitoring for at least 27 days. All emulsions showed similar results with a decrease in the backscattering with the aging time at the bottom and at the top of the vessel containing the sample, which is coherent with the existence of a destabilization mechanism by creaming and oiling off, respectively (McClements, 2015). There was no change in the backscattering in the middle of the measuring cell which demonstrated droplets sizes remained invariable (Mengual et al., 1999). This result is in agreement with that obtained from the laser diffraction technique. In Figure 3, the backscattering evolution over the whole of the container height as a function of the aging time is shown for one selected emulsion, IKA-SL5M. Additionally, in order to compare the different emulsions studied and better visualize the changes produced, in Figure 4 the backscattering in reference mode (ΔBS) of these emulsions at 27 days of aging time as function of the measuring cell length is shown. This figure supports the analysis above, namely all emulsions suffered similar effects, a marked decrease in the ΔBS at the bottom as a consequence of a creaming

process and a slight decrease in ΔBS at the top due to an oiling off destabilization phenomenon. This last phenomenon appears by coalescence of droplets of cream phase (Mengual et al., 1999) which provokes the occurrence of this oil layer at the top of the vessel.

The destabilization kinetic of the creaming process, the creaming index (IC) (McClements, 2007), as a function of the aging time has been calculated by the following equation (1):

$$CI(\%) = \frac{H_S}{H_E} \cdot 100 \quad (1)$$

where CI is the creaming index, H_S is the creaming layer length and H_E is the sample length. CI as a function of the aging time is plotted in Figure 5. The creaming rate, ω, has been determined from this figure by means of equation 2 (Trujillo-Cayado et al., 2017):

$$\omega = \frac{d(CI)}{dt} \cdot \frac{H_E}{100} \quad (2)$$

As can be observed in Figure 5, SL5M emulsion presented a lower slope and, as a consequence, a lower creaming rate ω= 0.182 %/day. This value contrasts, for example, with that obtained for IKA-SL5M and UT50 emulsions which showed a creaming rate of 0.218 and 0.698 %/day, respectively, being this last obtained from the linear part of the curve, that is to say, during the first 200 hours of study. This fact reveals SL5M emulsion showed the lowest destabilization due to creaming. It must be taken into account that the destabilization by creaming is strongly affected by the medium viscosity and the size and polydispersity of the droplets. In these emulsions, large sizes and span values are reached, as is supported in Table 1.

To quantify all the possible destabilization processes that occur in the emulsions simultaneously, a parameter called "Turbiscan Stability Index" (TSI) obtained from equation (3) has been used (Xu et al., 2016):

$$TSI = \sum_i \frac{\sum_h |scan_i(h) - scan_{i-1}(h)|}{H} \quad (3)$$

Where $scan_i(h)$ is the average backscattering for each time (i) of measurement, $scan_{i-1}(h)$ is the average backscattering for the (i-1) time of measurement and H is the number of scans carried out on the sample. As can be deduced from this equation, a higher value of TSI means a lower stability.

The TSI values as a function of the aging time have been plotted in Figure 6. The lowest value of this parameter at 27 days is exhibited by the SL5M emulsion, this being, therefore, the most stable emulsion.

As a consequence of the results obtained in this section, the emulsion obtained using only the Silverson L5M device will be used as the primary emulsion, namely as the starting point, for the next study.

From the environmental point of view, interesting finding can be noted. On the one hand, the elimination of the premix stage leads to a substantial saving in energy consumption. And the other hand, this fact provokes an important reduction in washing water.

Influence of the pressure on the microstructure and physical stability of microfluidized emulsions

In this part of the work, the most stable emulsion obtained previously, SL5M, was used as the primary emulsion to feed a M110P Microfluidizer homogenizer and was submitted to different pressures. The pressures applied ranged from 5000 to 25000 psi.

Laser diffraction results were obtained 24 hours after their production and are shown in Figure 7. As can be observed in Figure 7 and Table 2, the droplet sizes of microfluidized

emulsions were much smaller than those processed only with a rotor-stator system, with or without premix.

The small sizes zone of droplet size distribution tends to move towards smaller diameters and, therefore, to decreased mean Sauter diameters as the applied pressure increases in the range of 5000-20000 psi. The emulsion prepared at 25000 psi had higher $D_{3,2}$ values than for the emulsion processed at 20000 psi. In addition, from 15000 psi the distributions become bimodal, being monomodal only in the range 5000-10000 psi. Furthermore, the high sizes zone of DSD shifts to higher values. As result of these facts, the mean volume diameter and span significantly increase at the highest pressures. This could be due to the fact that, from these pressures, although drops of smaller diameters are formed, an over-processing is produced in other droplets which causes recoalescence and, as a consequence, the formation of a distribution tending to larger sizes (García et al., 2016; Trujillo-Cayado et al., 2018).

Figure 8 shows, by way of example, the droplet size distribution as a function of the aging time for the emulsion obtained at 20000 psi. As can be observed, there was a slight displacement of the distribution toward higher droplets sizes. However, these differences were not significant, as can be deduced from the comparison of Sauter diameters and their standard deviation at 1 day and 14 days which were $D_{3,2} = 0.52 \pm 0.02$ μm and $D_{3,2} = 0.54 \pm 0.02$ μm, respectively. Additionally, Figure 9, where the physical stability is analyzed by multiple light scattering, exhibits only a slight variation in ΔBS in the middle of the vessel at 55 days, which again indicates that this slight coalescence or Ostwald ripening process was not important. Therefore, Figure 9 also demonstrates that there was no change in the droplet size. This statement can be extrapolated to all microfluidized emulsions. In contrast, the emulsions submitted to pressure exhibited destabilization by

creaming and oiling off similarly to the primary emulsion, despite having much smaller mean droplet sizes. In any case, the peak (decrease of ΔBS at the bottom) is of lower width for emulsions submitted to microfluidization. The higher study time for the latter emulsions (55 days against 27 days) should be noted. Regarding these emulsions, the one with the least destabilization by creaming (least intensity and width in the abovementioned peak) corresponds to the emulsion prepared at 20000 psi, which is the one with the lowest average Sauter diameter.

Figure 10 shows the creaming index versus aging time for every microfluidized emulsion. As can be observed, after a delay period, a linear dependence of the creaming index with time occurred. From the slope of this region, it is possible to obtain the creaming rate (ω) and, therefore, the gravitational separation resistance. Table 3 exhibits the creaming rate (ω) and the time of creaming onset ($t_{0,c}$). As can be observed in this table, the emulsions obtained at 15000 psi and 20000 psi exhibited the lower creaming rate, and therefore were the more stable emulsions against creaming, although among them, the most stable emulsion is the one processed at 20000 psi due to the fact that the creaming destabilization starts later. This result is in agreement to results obtained by laser diffraction. The more stable emulsions in relation to the creaming effect were those with smaller drop sizes (15000 and 20000 psi) (McClements, 2015). It should be noted that in all cases, the values of ω were much lower than the values obtained for the primary emulsion, the emulsion that did not have a stage of secondary homogenization at high pressure (SL5M).

In Figure 11 the TSI values are plotted against the aging time for the microfluidized emulsions. It is again shown that the most stable emulsion is that prepared at 20000 psi. This emulsion exhibited not only the lowest level of creaming but also the lowest level of oiling off, as illustrated in Figure 9. As can be previously observed, the emulsions

obtained at 25000 psi exhibited a faster destabilization by creaming process than those of 15000 psi and 20000 psi as a consequence of its higher droplet size. This fact was consistent with the results presented in Figure 11. It is noteworthy again that the values of TSI, in all cases, were much lower than those shown by the emulsions from the first part of the study, even taking into account that in these emulsions the aging time was much longer.

Conclusions

Concentrated O/W emulsions containing 40 wt% bitter fennel oil and a bio-derived surfactant (Appyclean 6548, 4 wt %) were obtained. From an environmental point of view, these emulsions exhibit several advantages: a) they are aqueous based formulations which facilitates its application b) they are concentrated which provides savings in the transportation c) they are formulated with friendly raw materials which causes that the resulting wastes are less harmful. All these facts contribute to reduce the impact on the environment. Along with the formulation, other point must be taken into account which is the production procedure. Firstly, the influence of the incorporation of a premix stage prior to homogenization on the droplet size distribution and physical stability of the emulsions was investigated. The results obtained showed that the use of a premix worsens the physical stability of the emulsions and increases the average droplet diameters as a consequence of recoalescence due to excessive mechanical energy and to the entry of pre-existing droplets into the rotor-stator system. This finding contributes to an important saving in the energy consumption and the washing water and, therefore, a more sustainable production. The best results were obtained for the emulsion prepared only with Silverson L5M which, at the same speed, consumes less energy leading to a more

efficient energy process. All the emulsions developed in the first part of the investigation had mean droplet sizes greater than one micron, as well as destabilization by creaming and oiling off. The creaming rate and the physical stability of the emulsions are directly related to the droplet sizes. For this reason, the most stable emulsion and the one that presented the least destabilization by creaming was that which had the lowest average diameters that obtained only by means of Silverson L5M, without premix. Subsequently, the influence of homogenization pressure (between 5000 and 25000 psi) on the Silverson L5M emuslion was studied in a high energy microfluidization system, Microfluidizer M110P. These emulsions had submicron droplet sizes, much lower than any of those obtained in the first part and, as a result of this, better physical stability. The mean diameters and the turbiscan stability index decreased as the pressure increased in the range of 5000-20000 psi. The emulsion processed at 25000 psi showed larger diameters and, as a consequence, worse physical stability, probably due to a recoalescence process induced by an excess of energy. Thus, the emulsion that had smaller droplet sizes, as well as greater physical stability, was that prepared at 20000 psi.

As above mentioned, this work contributes to the development of stable agrochemical prototypes which are sustainable. These emulsions are sustainable not only by their formulation, based on essential oil and wheat waste surfactant but also by their processing protocol which could be conductive to a reduction of the impact on the environment.

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Figure captions

Figure 1. Influence of the processing protocols on the DSD of the primary emulsions at 1 day of aging time. Room temperature.

Figure 2. Backscattering in whole measuring cell length as a function of aging time for IKA-SL5M emulsion.

Figure 3. Delta backscattering at 27 days of aging time as a function of the measuring cell length for all studied systems.

Figure 4. Influence of the processing protocol on the creaming index (CI) as a function of aging time.

Figure 5. Effect of aging time on the droplet size distributions for SL5M emulsion. Room temperature.

Figure 6. Influence of the processing protocol on the Turbiscan Stability Index as a function of aging time.

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Tables

Table 1. Influence of processing protocol on the mean diameters and span values. Room temperature.

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Table 3. Influence of the applied pressure on the creaming rate (ω) and the time of creaming onset ($t_{0,c}$).

Table 1. Influence of processing protocol on the mean diameters and span values. Room temperature.

	$D_{3,2}$ (μm)	$D_{4,3}$ (μm)	Span
IKA - UT50	2.37 ± 0.07	2.91 ± 0.09	1.215 ± 0.068
UT50	2.04 ± 0.05	2.63 ± 0.07	1.079 ± 0.051
IKA - SL5M	1.98 ± 0.05	2.57 ± 0.07	0.988 ± 0.044
SL5M	1.82 ± 0.06	2.29 ± 0.05	1.164 ± 0.071

Table 2. Influence of processing protocol on the mean diameters and span values. Room temperature.

Pressure (psi)	$D_{3,2}$ (μm)	$D_{4,3}$ (μm)	Span
SL5M	1.82 ± 0.06	2.29 ± 0.05	1.164 ± 0.071
5000	0.74 ± 0.04	1.06 ± 0.06	1.541 ± 0.088
10000	0.68 ± 0.03	1.17 ± 0.07	1.295 ± 0.065
15000	0.59 ± 0.02	0.91 ± 0.05	2.042 ± 0.101
20000	0.52 ± 0.02	1.53 ± 0.08	4.585 ± 0.327
25000	0.78 ± 0.03	1.70 ± 0.08	3.506 ± 0.287

Table 3. Influence of the applied pressure on the creaming rate (ω) and the time of creaming onset ($t_{0,c}$).

Pressure (psi)	ω (%/day)	$t_{0,C}$ (h)
5000	0.054	0
10000	0.053	125
15000	0.031	355
20000	0.030	436
25000	0.039	136

Figure captions

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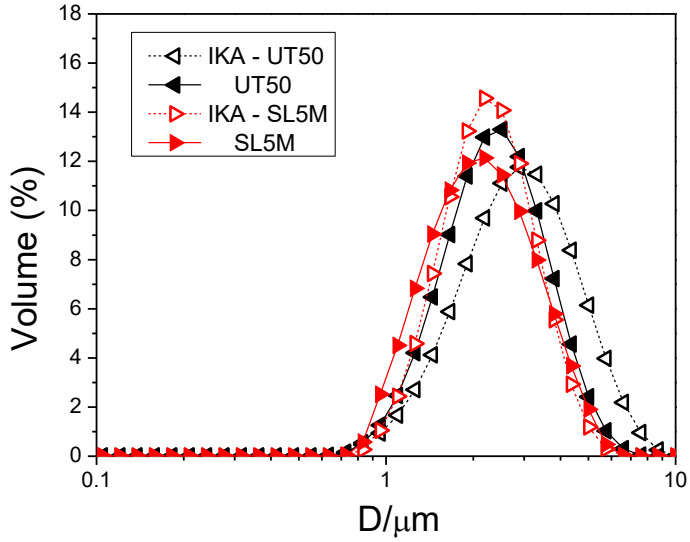

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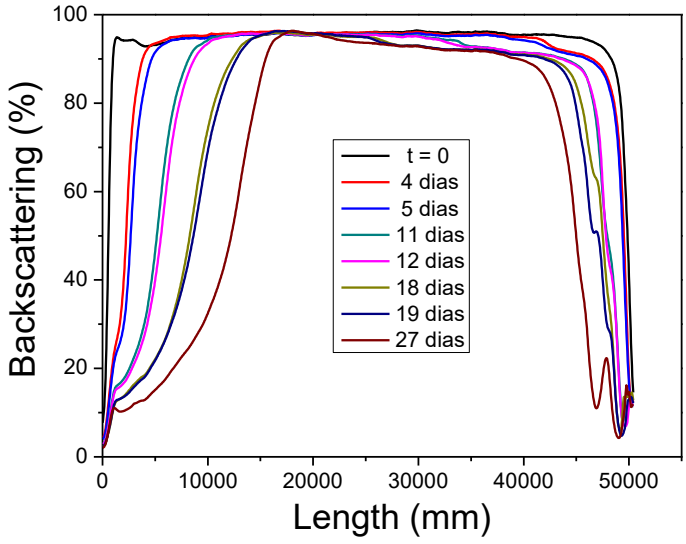

Figure 2. Backscattering in whole measuring cell length as a funtion of the aging time for IKA-SL5M emulsion

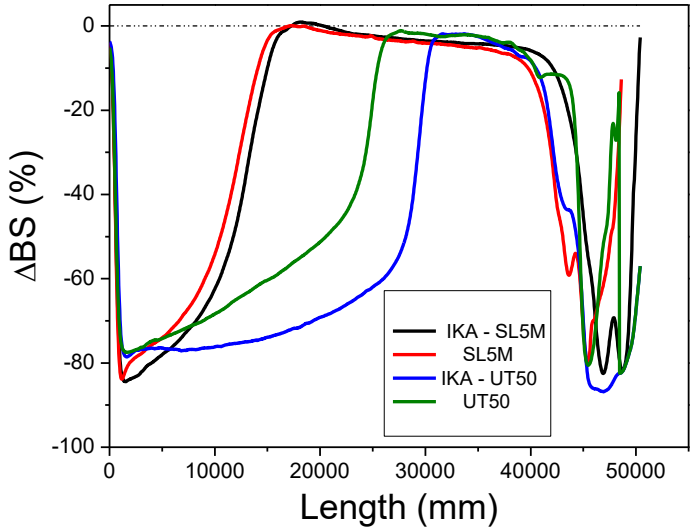

Figure 3. Delta backscaterring at 27 days of aging time as a function of the measuring cell length for all studied systems

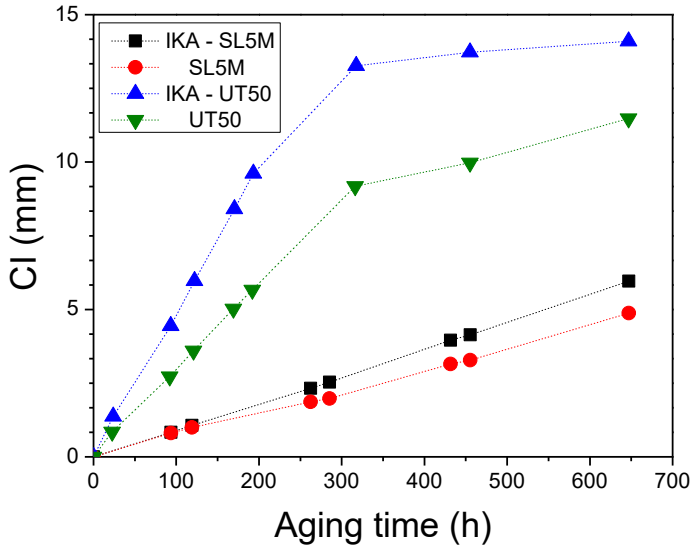

Figure 4. Influence of the processing protocol on the creaming index (CI) as a function of the aging time.

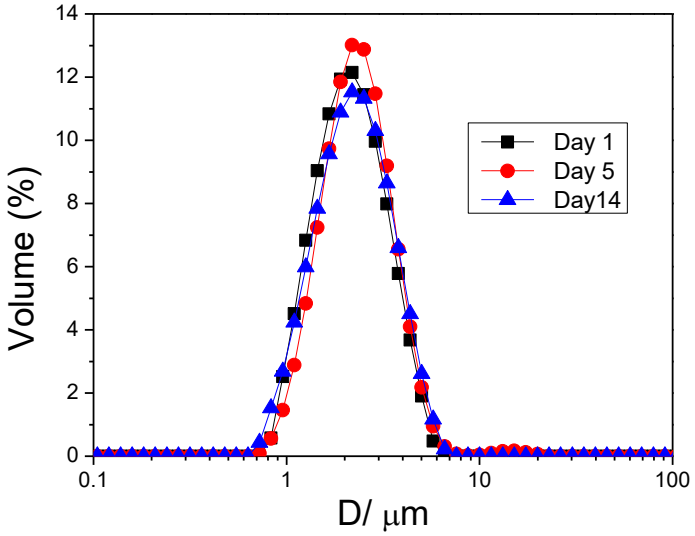

Figure 5. Effect of the aging time on the droplets sizes distributions for SL5M emulsion. Room temperature

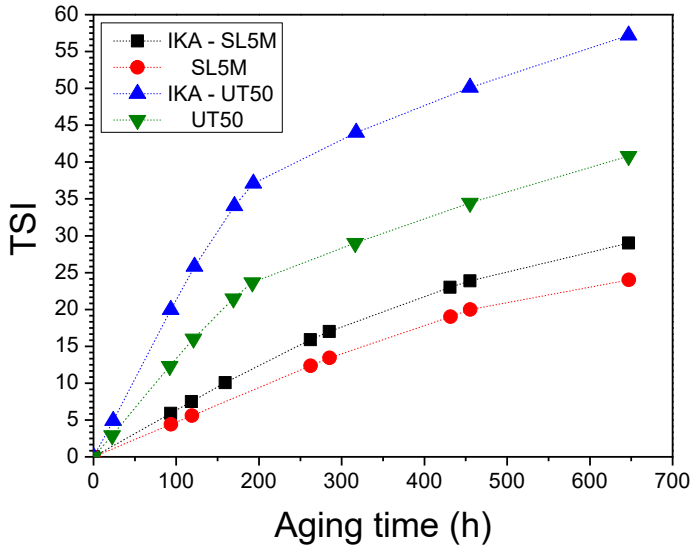

Figure 6. Influence of the processing protocol on the Turbiscan Stability Index as a function of the aging time.

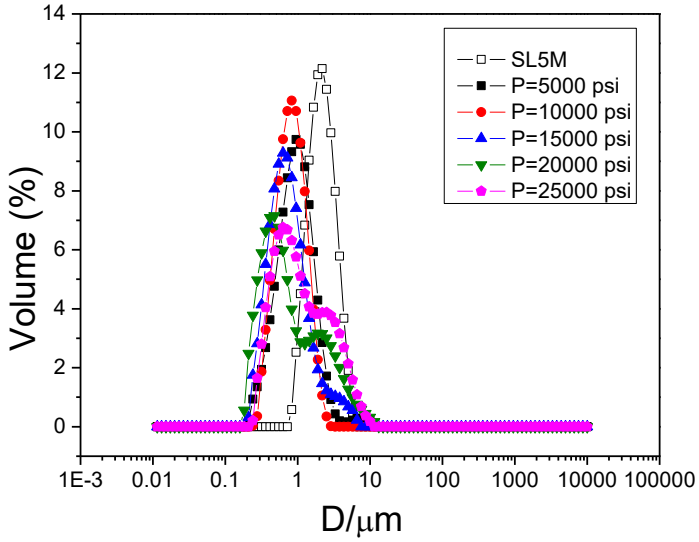

Figure 7. Influence of the pressure applied by M-110P microfluidizer homogenizer on the droplets sizes distribution. Room temperature.

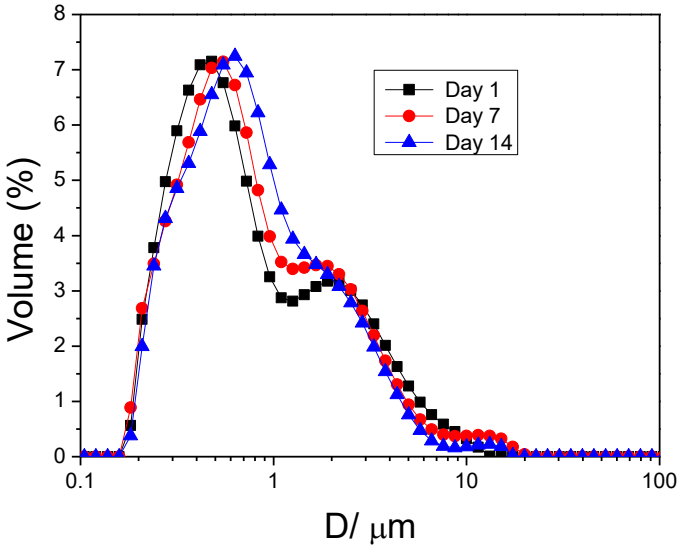

Figure 8. Influence of aging time on the droplet size distribution of the emulsion obtained at 20000psi.

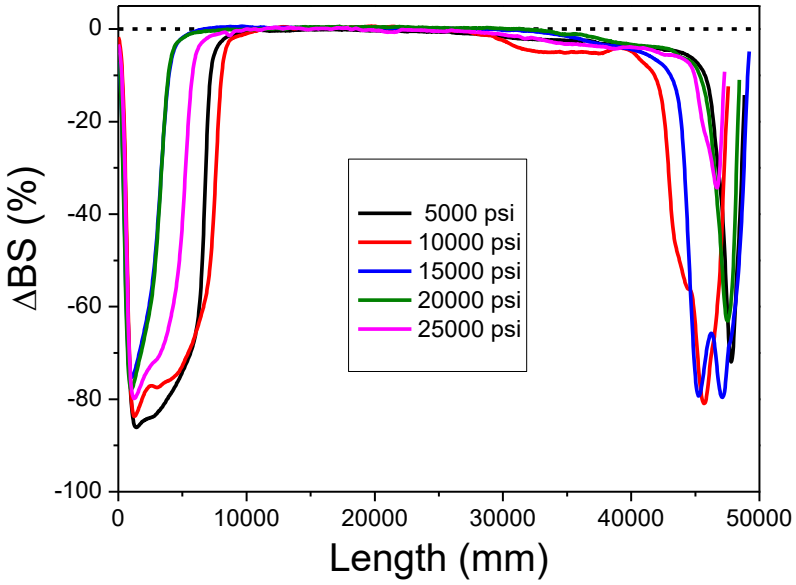

Figure 9. Influence of the applied pressure on th ΔBS as a function of the measuring cell length at 55 days of aging time.

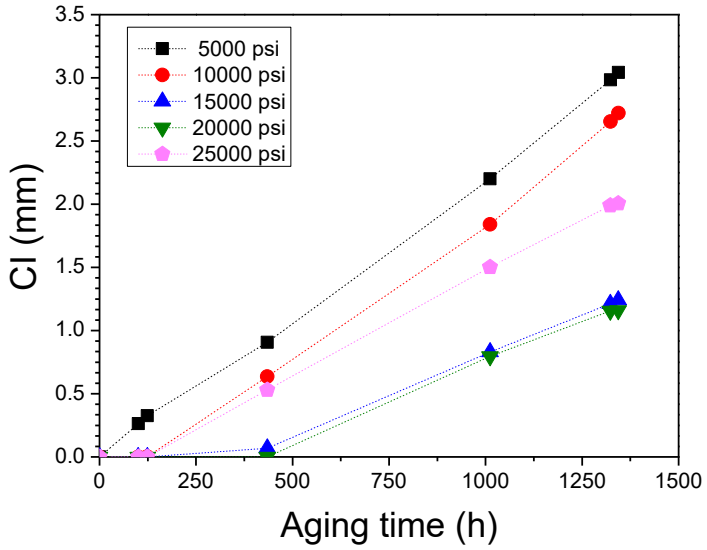

Figure 10. Influence of the applied pressure on the creaming index as a function of the aging time.

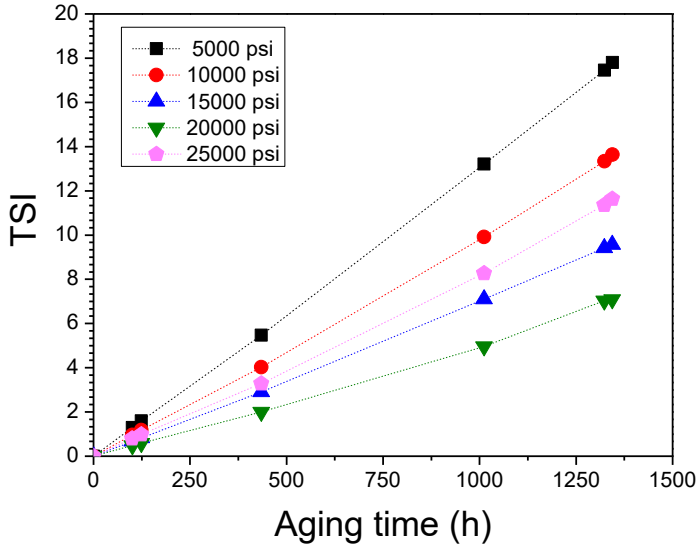

Figure 11. Influence of applied pressure on the TSI as a function of the aging time