Preparation of monodisperse O/W emulsions using a crude surface-active extract from argan by-products in microchannel emulsification

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Abstract

In this study, we evaluated the performance of a mildly derived extract from argan by-products in microchannel emulsification (MCE). Our aim is to produce stable monodisperse O/W emulsions using this extract as a sole emulsifier. Preliminary investigations about the characteristics of argan extract indicated the potential interaction between surface-active components (i.e. saponins, proteins), in bulk phase prior to adsorption at the oil/water interface, resulting in the formation of biogenic complexes with strong interfacial properties. This is important for successful MCE as this technique depends exclusively on dynamic interfacial tension reduction for droplet formation. Nevertheless, upon performance of emulsification experiments, we also found that the complex composition of this extract could counteract its emulsifying efficiency, by creating a hydrophobic, or slightly hydrophilic, layer on the MC array plate surface. This resulted in unsuccessful emulsification using short MCs but did not affect the emulsification behavior in longer ones. Using longer MCs, we could produce stable monodisperse O/W emulsions, with similar droplet size (~ 36 µm) and droplet size distribution (Relative span factor < 0.25) to those obtained using Tween 80, and for up to 10 h of continuous emulsification.

Keywords: Argan by-products; crude emulsifying extract; monodisperse emulsion; microchannel emulsification.

1. Introduction

Emulsifiers are amphiphilic molecules or particles, capable of adsorbing at the hydrophile-hydrophobe interface, thus reducing the tension between immiscible liquids. Due to their surface-activity and versatile properties, emulsifiers have various applications over a broad range of industries. Particularly, they are used as stabilizers and/or encapsulants in food, cosmetic and pharmaceutical products, in which they provide multiple properties such as good dispersibility, prolonged stability and improved bioavailability of other ingredients [1].

For many years, the emulsifiers industry has established various processes for the production of emulsifying compounds, with exhibited properties and competitive prices. Consequently, many of the emulsifiers used nowadays still derive from the same chemical and/or enzymatic reactions that were primarily designed in the past for mass and economic production of these substances [2]. The current trend however towards natural and sustainable products have necessitated manufacturers to find natural alternatives to synthetic emulsifiers. As a result, intensive research has been done to extract, identify and characterize various natural emulsifiers from different origins [3].

Proteins, phospholipids, and polysaccharides have been successfully obtained previously and evaluated for their emulsifying/stabilizing properties [4]. However, despite being suitable for multiple applications, many of these emulsifiers are criticized, particularly because of their intensive extraction and/or fractionation/purification methods [5, 6]. A new trend in the utilization of natural emulsifiers suggests, therefore, the use of less intensive practices for a sustainable production of emulsion-based products. Particularly, the use of crude plant

66 emulsifying extracts, obtained via simple extraction/separation steps, have been largely
67 considered in recent times.

68 Ralla et *al*. [7] evaluated the emulsifying properties of a crude surface-active extract from red
69 beet by-products. They found that concentrations as low as 0.75% (w/w) of this extract could
70 produce stable oil-in-water (O/W) emulsions using high-pressure homogenization. Other
71 findings also include the good emulsifying properties of mildly derived extracts from oat bran,
72 mullein, and sunflower seeds [8, 9, 10]. In all these cases, however, emulsions were prepared
73 using high-energy emulsification methods to increase the chances of obtaining stable emulsions.
74 On the other hand, the effect of multicomponent surface-active extracts on the performance of
75 low-energy emulsification methods such as microchannel emulsification (MCE) is still unknown.

76 MC is a microfluidic emulsification method that enables the production of monodisperse
77 emulsions using microfabricated channels, embedded on a silicon chip [11]. It is particularly
78 promising for the encapsulation of bioactive compounds in O/W emulsions, as it can provide
79 superior physicochemical stability to the prepared emulsions [12]. The technique is based on a
80 simple process that involves no mixing or shear processing, as compared to conventional
81 emulsification methods. Briefly, the dispersed phase is pressurized through the channels and the
82 droplets are spontaneously formed by the effect of interfacial tension (in typically less than 0.01
83 s). During this time, the emulsifier should rapidly adsorb onto the oil/water interface to prevent
84 newly generated droplets from coalescence [13]. Therefore, factors such as the speed of
85 adsorption of surface-active components at the oil/water interface, their extent at reducing of
86 interfacial tension, as well as their ability to spontaneously form a continuous film around the
87 droplets, are crucial in MCE [14].

In this study, we evaluated the emulsifying performance of a surface-active extract from argan by-products in MCE. Our aim is to produce stable monodisperse O/W emulsions using this extract as a sole emulsifier.

2. Materials and methods

2.1. Materials

Argan by-products refer to the press-cake obtained after oil extraction. The cake was provided by an oil-producing cooperative in the region of Essaouira, Morocco, and stored at -20 °C until use. Ethanol (99.5%), acetonitrile (HPLC grade), formic acid (HPLC grade), refined soybean oil and polyoxyethylene (20) sorbitan monooleate (Tween 80) were purchased from Wako Pure Chemical Industries, Ltd. (Osaka, Japan). Whatman filter paper (grade 1) was obtained from GE Healthcare Life Sciences (Chicago, Illinois, USA). Disposable syringe filters (0.22 μm pore size, Nylon) were obtained from Advantech Co., Ltd. (Taipei, Taiwan). Ultrapure water with a resistivity of 18 MΩ·cm was produced by an ultrapure water system (Arium® pro, Goettingen, Germany) and used to prepare all solutions in the current study.

2.2. Preparation of argan extract

Argan press-cake was initially ground into small particles (< 0.5 mm) using a high-speed mixer. The obtained powder (100 g) was then added to a fresh mixture (500 mL) of water and ethanol (20/80, v/v) and extracted for 2 hours at room temperature. After centrifugation (2000 g for 30 min) and filtration (Whatman paper) to remove solid particles, the solvent was then evaporated under reduced pressure to give a syrupy residue that was re-dispersed in water and re-filtered using a 0.22 μm syringe filter. Finally, for an accurate preparation of the working solutions and to prevent microbial growth during storage, the extract was freeze-dried for 2 days at -80 °C and 5 Pa and kept at 5 °C until use.

2.3. *Physicochemical characterization of argan extract*

Total proteins and saponins contents were previously described in Taarji et *al*. [15]. Herein, the presence of glucuronic-type saponins was evaluated by means of LC/MS analysis (LCMS-8050, Shimadzu Ltd., Kyoto, Japan), following the method of Henry et *al*. [16]. Briefly, a 10 µL aliquot of argan extract was injected into a C18 separation column (250 x 2.1 mm^2, 5 µm, Shim-pack, Shimadzu Ltd., Kyoto, Japan) and the compounds of interest were eluted using a linear 25 min gradient, from 30 to 60% acetonitrile in 0.1% formic acid, at 0.4 mL/min and 50 °C. The mass spectrometer operated then in the negative ion electrospray ionization mode ESI(-), acquiring a full scan from 1000-1500 m/z.

Viscosity was measured using a vibro-viscometer at 25 °C (SV-10, A&D Co., Ltd., Tokyo, Japan). Particle size and surface charge were measured using a Dynamic Light Scattering/ζ-potential Analyzer (Nano ZS, Malvern Instruments Ltd., Worcestershire, UK). Contact angles were measured according to the method of Zhang et *al*. [17], by measuring the angle of generated emulsifier drops to the plasma oxidized MC plate surface (PD-W, Kyowa Interface Science Co., Ltd., Saitama, Japan).

2.4. *Preparation of O/W emulsion using MCE*

2.4.1. *Microchannel array chips*

Asymmetric straight-through silicon MC array chips with different channel dimensions were used in this study. Figures 1a and 1b depict the structure of WMS 1-2 and WMS 1-3 chips, respectively. Both chips measure 24 x 24 x 0.5 mm^3 and include a 100 µm etched active area, containing MCs. Each MC consists of a 10 x 70 $µm^2$ cylindrical microhole on the bottom side and a rectangular microslot on the top side. However, WMS 1-3 contains 23 348 MCs, while WMS 1-2 contains only 10 313 MCs. This is because the microslots in WMS 1-2 (10 x 100 x 30

136 μm^3) are wider than those in WMS 1-3 (10 x 50 x 30 μm^3), which results in fewer channels on
137 the horizontal line. In WMS 11-1 (Figure 1c), the active area is etched to only 200 µm, resulting
138 in longer microholes on the bottom side (160 µm). Here, each MC (13 752 MCs) consists of a 10
139 x 160 μm^2 microhole and a 10 x 80 x 40 μm^3 microslot.

140 Before conducting the experiments, the chips were ultrasonicated at 100 KHz with neutral
141 detergent, disinfectant ethanol, and ultrapure water, respectively. They were then subjected to
142 plasma oxidation (PR500, Yamato Science Co. Ltd., Tokyo, Japan), to develop a hydrophilic
143 silicon dioxide (SiO_2) layer on the surface. For the rest of the time, they were stored in water at
144 room temperature.

145 *2.4.2. Emulsification procedure*

146 Emulsions were prepared at room temperature as illustrated previously by Khalid et *al.* [18].
147 First, the MC chip was mounted into the emulsification module, with the microslots facing the
148 top. Next, soybean oil was injected to the bottom side of the chip, using a programmable syringe
149 pump (Model 11 Elite, Harvard Apparatus Inc., Holliston, USA), at 2 mL h^{-1}. When the droplets
150 started to generate, the continuous phase (1%, w/w) was finally delivered, from an elevated
151 reservoir to the top side of the chip, which enables the recovery of generated droplets. We
152 decided, to fix the dispersed phase flow rate at 2 $mL.h^{-1}$, to allow easier visualization of droplet
153 generation, because higher flow rates resulted in rapid generation.

154 *2.5. Droplet characterization*

155 During emulsification, the droplet generation behavior was recorded using a CCD microscope
156 video system (MS-511B, Seiwa Kougaku Sesakusho Ltd., Tokyo, Japan), equipped with 2.5 or 5
157 magnification objective lenses. After emulsification, the droplet size was measured using a laser

158 diffraction particle size analyzer (LS 13,320, Beckman Coulter, Brea, USA) and the droplets
159 were observed using an optical microscope (Leica DFC 7000 T, Wetzlar, Germany). The average
160 droplet size was expressed as volume mean diameter ($d_{4,3}$), while the droplet size distribution
161 was presented by relative span factor (RSF).

162 **3. Results and discussion**

163 *3.1. Physicochemical characteristics of argan extract*

164 Argan extract was prepared using a simple solvent extraction procedure, without fractionation or
165 purification of the obtained composition. It is, therefore, a mixture of multiple compounds that
166 could mutually affect the surface-active and/or emulsifying properties, as compared to individual
167 ingredients. Earlier investigation about the composition of this extract indicated the presence of a
168 significant amount of triterpenoid saponins (~ 3.5% (w/w)) [15]. These compounds have an
169 amphiphilic chemical structure and a relatively low molecular weight, thus acting as small
170 molecule surfactants [3]. The use of triterpenoid saponins has been shown previously to reduce
171 the interfacial tension and to produce stable O/W emulsions. Nevertheless, a relatively high
172 concentration of saponins is usually required to produce stable emulsions [19]. Recent
173 applications of saponins in O/W emulsions, suggest therefore their use in mixed surfactant
174 systems, for improved surface-active and/or emulsifying properties. Reichert et *al*. [20] for
175 example found that the concentration of Quillaja saponins could be successfully reduced, without
176 changes in emulsion's droplet size or droplet size distribution, following the addition of limited
177 amounts of sodium caseinate. Other findings also include the synergetic effect of saponins and
178 proteins in reducing the interfacial and surface tensions [21]. Therefore, it is suggested that the
179 presence of saponins and proteins in mixed surfactants systems results in optimal molecular
180 complexes that enhance their adsorption at the oil/water or air/water interfaces [22, 23].

In our previous study, we also identified the presence of a lower concentration of proteins in the present extract (~ 1% (w/w)) [15]. However, it was not clear how the mutual presence of saponins and proteins could result in the formation of biogenic complexes that could explain the strong interfacial properties (Table 1). In literature, the molecular complexation of saponins and proteins is usually explained by the action of low molecular forces such as hydrogen bonds, hydrophobic and electrostatic interactions. Wojciechowski et *al*. [24] for example suggested that the presence of deprotonated glucuronic acid in the molecules of Quillaja saponins could result in electrostatic interactions with the positively charged domains in lysozyme proteins. Similar behavior was also suggested in Quillaja saponins/β-casein and Quillaja saponins/β-lactoglobulin systems [21, 23]. In the present extract, we identified several glucuronic-type saponins in argan extract (Figure 2), by comparison to the previously obtained ESI(-)-MS chromatograms by Henry et *al*. [16]. These compounds have an acidic pKa of around 3.25 and therefore tend to be completely deprotonated at neutral pH [3], which can result in electrostatic interactions with proteins.

The formation of proteins/saponins complexes may occur either in bulk solution, prior to adsorption, or directly at the oil/water interface, through co-adsorption of the slower diffusing component on the pre-adsorbed layer of the other component [24]. To gain more insight into the structural organization of surface-active components in our extract, we measured the particle size and the particle size distribution of the prepared solutions. As shown in Table 1, argan extract has an average particle size of around 133 nm, while Tween 80 resulted in a smaller size of 9.4 nm. Normally, small molecule surfactants such as Tween 80 and saponins are expected to result in smaller micellar structures, in comparison to proteins or proteins/saponins complexes. Mitra & Dungan [25] for example found that the particle size of pure Quillaja saponins solutions

204 generally ranges from 6-12 nm. Tween 80 micelles size should be also in that range due to their
205 small molecule structure. In our extract, however, the particle size measurement resulted in a
206 single peak that starts from about 50 nm (Figure 3). This means that no smaller micellar
207 structures were present in this system and therefore saponins were exclusively adsorbed on larger
208 aggregates or present in free soluble form.

209 Evidence presented in this study indicates that argan extract contains relatively large aggregates,
210 which can result from electrostatic interactions between glucuronic-type saponins and proteins.
211 Interestingly, the ς-potential measurement of this extract indicated a strong negative charge of
212 about -51 mV (Table 1). It is difficult to determine the exact source of this charge. However,
213 since ς-potential is the measurement of the surface charge of colloidal particles in dispersion, we
214 correlate this charge to the previously identified aggregates in argan extract. Normally, upon
215 depletion of the positively charged domains in the protein structures, we can also suggest the
216 formation of other types non-covalent bindings with saponins. Böttcher et *al*. [22] for example
217 hypothesized that in binary systems, saponins and proteins can interact together by hydrogen
218 bonds and/or hydrophobic interactions, resulting in the formation of biogenic complexes.
219 Therefore, in our speculation, the strong negative charge in argan extract could be due to the
220 glucuronic-type saponins that are non-electrostatically adsorbed to the complex aggregates
221 and/or to the negatively charged domains in the protein structures themselves. Actually, in our
222 previous study, we also found that emulsions prepared using a similar extract from argan press-
223 cake, by high-pressure homogenization, were sensitive to extreme acidic pH, resulting in the loss
224 of droplets' negative charge and physical stability of emulsions [15]. However, even at pH 3,
225 providing that the pKa of glucuronic-type saponins is around 3.25 [3], the emulsion droplets still

had a relatively high negative charge on their surface, which indicates that saponins might not be the only contributors to this charge.

Overall, the composition and interfacial activity of argan extract seems to be favorable for successful MCE. The formation of proteins/saponins complexes is possible and this is believed to provide strong emulsifying properties to this extract. Other factors, such as the low viscosity and the relatively small particle size (Table 1) are also believed to acquire a smooth droplet generation during MCE, by allowing a fast inflow of the continuous phase, as compared to the outflow of the oil phase [26].

3.2. Droplet generation behavior in MCE

MCE was introduced to the first time by Kawakatsu et *al*. [27] as a microfluidic technique for the preparation of monodisperse emulsions. Since that time, various MC designs and geometric structures have been fabricated and evaluated for their efficiency using this technique. Nowadays, asymmetric straight-through MC arrays are the most commonly used devices in MCE. They are particularly robust at carrying stable droplet generation behaviors using high dispersed phase flow rates and low viscosity oils, hence being more promising for mass and economic production of monodisperse emulsions [12]. In this section, we used three different asymmetric straight-through MC array chips, with various microhole lengths and microslots widths, to evaluate the emulsifying behavior of argan extract in MCE. Live visualization of droplet formation was possible thanks to the transparency of argan extract and Tween 80 continuous phases (Figure 3b). Video records are provided in supplementary materials; the corresponding stills are presented hereafter.

As shown in Figure 4, droplet generation in WMS 1-2 and WMS 1-3 was not successful using argan extract as emulsifier. The dispersed phase that passed through the working channels started

to inflate rapidly along the plate surface until neighboring droplets contacted each other and coalesced into larger particles. At this point however, it was not clear whether this behavior results from a limited concentration of the emulsifying compounds in the system, from the interaction between the plate surface and the extract composition or simply due to a defect in the MCs design. We conducted, therefore, new experiments using longer MCs and compared the droplet generation behavior to the commonly used surfactant Tween 80. As shown in figure 4, Tween 80 exhibited a smooth droplet generation behavior, regardless of the MCs dimensions. On the other hand, argan extract was only successful in WMS 11-1, provided that the same concentration (1%, w/w) was used in all experiments.

During straight-through MCE, the oil phase is pressurized through the microholes, expended inside of the microslots and finally detached at the outlet of the microslots [28]. The pressure difference between the expended oil and the disrupted droplets plays an important role in this process [29]. This pressure difference can be estimated by Fanning's equation:

$$\Delta P = 4f \left(\frac{l}{deq}\right)\left(\frac{1}{2}\rho U^2\right) \qquad (1)$$

where ΔP is the pressure difference in the channel, f is Fanning's friction factor, deq is the equivalent diameter of the channel, ρ is the liquid density, U is the characteristic velocity, and l is the channel length [30]. As shown by equation 1, longer MCs produce larger pressure drops in the channel. Large pressure drops enhance the flow of the dispersed phase, thus allowing rapid droplet detachment [29]. Tween 80 allowed a smooth droplet generation when using short MCs, while argan extract was only functioning efficiently when using longer MCs. We therefore believe that the complex composition of argan extract disturbed the smooth flow of the oil phase,

inside of the MCs. Consequently, a higher pressure-drop was necessary to enhance droplet detachment.

Saito et *al*. [14] evaluated the emulsifying performance of various proteins in MCE. They found that when the pH was lower than the isoelectric point of the protein, electrostatic attractive forces worked between the negatively charged silicon MC plate and the positively charged protein molecules, resulting in unsuccessful emulsification. In our case, we previously showed that argan extract contains negatively charged particles (Table 1) that should repulse with the surface-oxidized SiO_2 surface of MCs. However, upon measurement of contact angles, we found that argan extract interacts more with the MC chip surface, in comparison to Tween 80 (Table 1). This means that other components of this extract have a higher affinity to the chip material, thus resulting in higher contact angles. In general, a higher contact angle means that the emulsifier solution yields more hydrophobic, or less hydrophilic, layer. A lower contact angle, on the other hand, means that the previously oxidized chip was less affected by the emulsifier solution composition, thus maintaining its original hydrophilic properties [17]. In the present extract, it was difficult to determine the exact compounds that could interact with the MC array chip surface. However, since the contact angle was increased (52.4°), as compared to Tween 80 (19.1°), we speculate that these compounds have an amphiphilic structure, with the polar regions binding to the silicon dioxides on the chip surface and the hydrophobic regions facing the chip exterior, thus creating a hydrophobic layer on the surface.

Tong et *al*. [31] and Kobayashi et *al*. [32] initially emphasized the importance of hydrophilic MC array plates for a successful production of monodisperse O/W emulsions using MCE. Consequently, plasma-oxidized chips are normally used to produce O/W emulsions, to avoid wetting of the oil phase on the chip surface. Nevertheless, it has been shown previously that the

293 slightest reduction in the hydrophilicity of the chip could greatly affect the particle formation
294 behavior in MCE [14, 17]. Using argan extract, this resulted in unsuccessful emulsification using
295 short MCs in WMS 1-2 and WMS 1-3. This malfunctioning, however, could be easily prevented
296 by using longer channels in WMS 11-1, providing stable monodisperse O/W emulsions with
297 similar droplet size (~ 36 µm) and droplet size distribution (Relative span factor < 0.25) to those
298 obtained by the commonly used surfactant Tween 80 (Figure 5).

299 *3.3. Long-term production stability of O/W emulsions*

300 The previous observations in section 3.2 indicate that argan extract interacts more with the MC
301 array chip by creating a hydrophobic, or less hydrophilic, layer on the chip surface. This resulted
302 in unsuccessful emulsification using WMS 1-2 and WMS 1-3 chips but did not affect the droplet
303 generation behavior in longer MCs of WMS 11-1. In this section, we evaluated the long-term
304 production stability of O/W emulsions prepared using argan extract, to assess the possible
305 deposition of such a layer over time, thus counteracting droplet formation.

306 As shown in figure 6a, the droplet size was unchanged over the entire period of emulsification.
307 The droplets were uniformly sized and spherical and were easily swept away by the continuous
308 phase cross-flow, without interaction with the chip surface (Figure 6b). Tong et *al*. [33] initially
309 demonstrated that stable droplet generation could be successfully maintained for up to 4 h when
310 using MCE. Vladisavljević et *al.* [34] also reported no change in droplet generation
311 characteristics after 7 h of continuous emulsification. In general, it seems that MCE is efficient
312 for the long-term production of monodisperse O/W emulsions. However, it has been shown
313 previously that the choice of emulsifier plays a key factor in continuous operating of MCE.
314 Tween 20, for example, was found to gradually increase the affinity between the oil phase and
315 the chip surface, thus resulting in variations of the droplet generation behavior and the droplet

316 size distribution over time. SDS, on the other hand, allowed a successful continuous
317 emulsification, by providing strong repulsive forces between the generated droplets and the chip
318 surface [35]. To assess the possible interaction between the MC chip surface and the generated
319 droplets using argan extract, we measured the ς-potential of prepared emulsions. As expected,
320 emulsions prepared using argan extract presented a highly negative droplet charge of around -70
321 mV (data not shown), in accordance with the previously identified particles (Table 1). We
322 suggest, therefore, that during emulsification using argan extract, the generated droplet repulsed
323 strongly with the MC array chip surface, due to the negatively charged coating provided by
324 surface-active components of this extract. Consequently, this allowed to maintain a smooth
325 droplet generation behavior over the entire period of emulsification, without variation in droplet
326 size or droplet size distribution. Meanwhile, long MCs in WMS 11-1 were necessary for a
327 successful droplet formation to occur in the first place, by creating a large pressure drop between
328 the channel chamber and the chip surface, thus facilitating the droplet detachment on the outer
329 edge of the channels.

330 **4. Conclusions**

331 This study demonstrated the potential application of crude surface-active compositions, such as
332 argan extract, for the preparation of monodisperse O/W emulsions by MCE. Unlike purified
333 emulsifiers, argan extract is a mixture of multiple compounds that can mutually affect emulsion
334 preparation. The surface-active component of argan extract can sufficiently cover newly
335 generated interfaces, thus enhancing droplet stability. In contrast, it can significantly reduce MC
336 chip surface hydrophilicity, thus disturbing droplet formation. Using longer MCs was necessary
337 for successful MCE using argan extract as emulsifier, by creating a high pressure-drop inside of
338 the channels, thus promoting droplet generation. In the future, it would be interesting to evaluate

this behavior using various emulsifying extracts from different origins. It would be also interesting to investigate other parameters such as the physicochemical stability of the prepared emulsions.

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Conflicts of interest

The authors declare no conflict of interest relating to the experimental results presented herein.

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Figures caption

Figure 1. Cross-sectional representation of MC array chips used in this study: (a) WMS 1-2, (b) WMS 1-3 and (c) WMS 11-1. Variable dimensions (μm) are highlighted in yellow.

Figure 2. Chemical representation of glucuronic-type saponins identified in argan extract. (a) m/z = 1251.6 (b) m/z = 1237.6 (c) m/z = 1235.6 and (d) m/z = 1221.6.

Figure 3. (a) Particle size distribution of aqueous phases containing argan extract or Tween 80 (1%, w/w) (b) Visual appearance of freeze-dried and freshly prepared aqueous phases of argan extract and Tween 80 (1%, w/w).

Figure 4. Effect of emulsifier type (1%, w/w) on the droplet generation behavior in (a) WMS 1-2, (b) WMS 1-3 and (c) WMS 11-1; bar = 200 μm. Stills were taken using a 2.5 magnification objective lens to show a larger area of the MC array plate and to highlight the oil inflation behavior over multiple channels. Video records (x5), showing the shape of the droplets and the speed of generation, are provided in supplementary materials.

Figure 5. (a) Droplet size distribution and (b) optical micrographs of emulsions (WMS 11-1) prepared using argan extract or Tween 80 (1%, w/w); bar = 40 μm. Measurements and microscope images were taken after 24 hours of storage at room temperature.

Figure 6. (a) Droplet size ($d_{4,3}$) and (b) droplet generation behavior of emulsions prepared using argan extract (1%, w/w), after 1 and 10 h of continuous emulsification (WMS 11-1); bar = 200 μm. Samples for droplet size measurement and video captions (x5) were taken within 5 min of appropriate emulsification time.

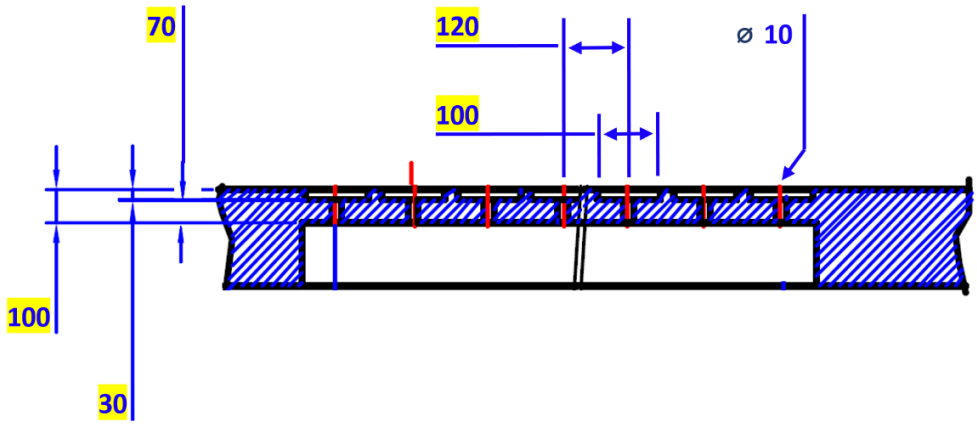
Table 1. Fluid properties of aqueous phases, containing argan extract or Tween 80 (1%, w/w).

	Argan extract	**Tween 80**
Interfacial Tension (mN m^{-1})	11.5 ± 0.3*	5.8 ± 0.3
ς-potential (mV)	-51.1 ± 2.0	-4.3 ± 1.6
Particle size (nm)	133.9 ± 0.5	9.4 ± 0.1
Contact angle (°)	52.4 ± 4.2	19.1 ± 2.1
Viscosity (mPa s)	0.95 ± 0.2	0.91 ± 0.0

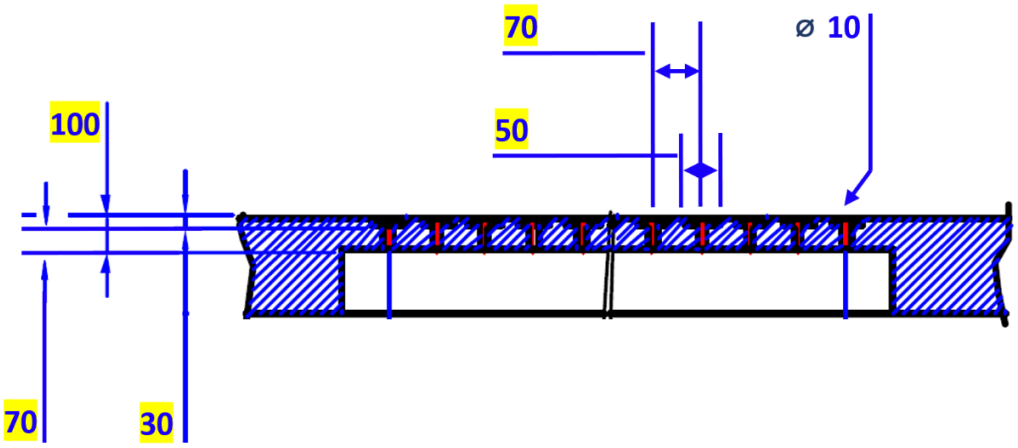
***Interfacial tension at the soybean oil/water interface [15].

Figure 1

(a)



(b)



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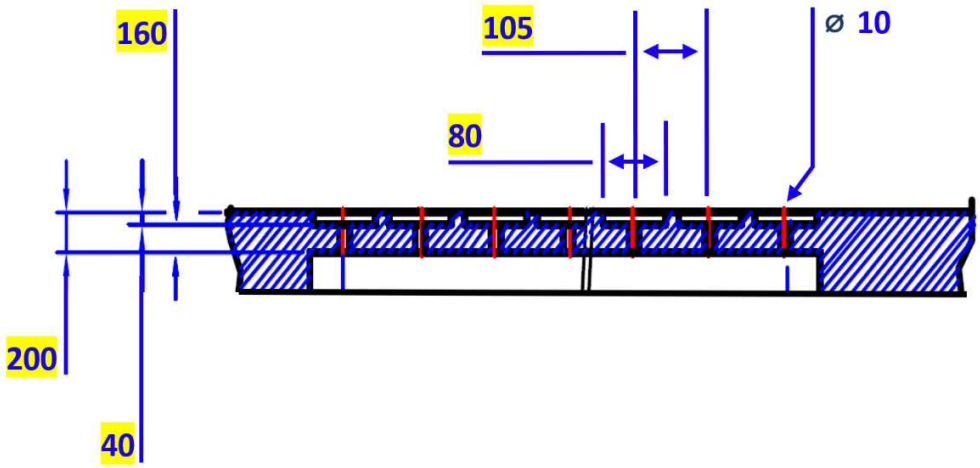
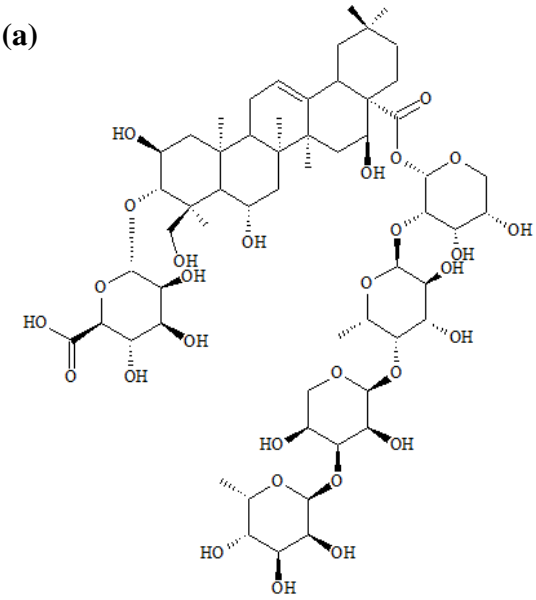


15 **Figure 2**

16

(a)



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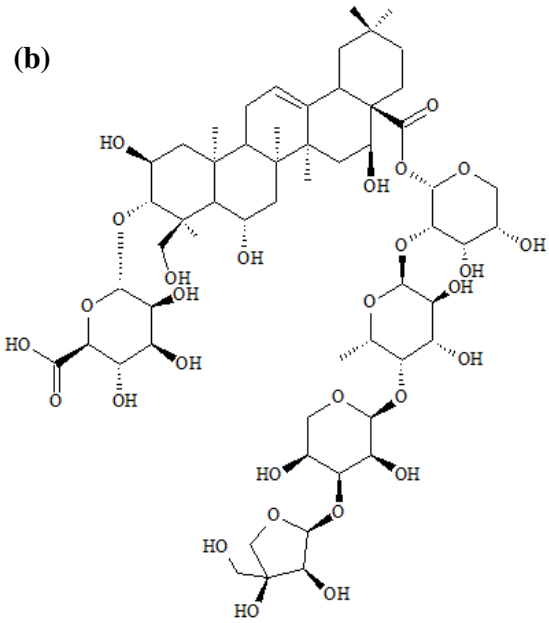

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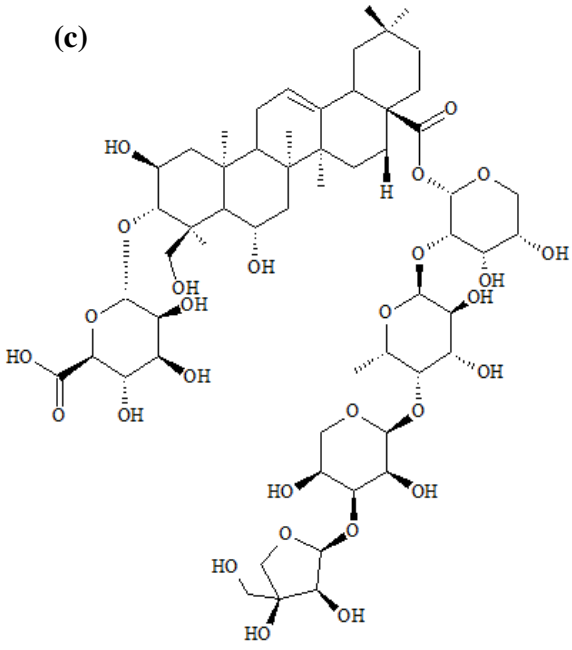
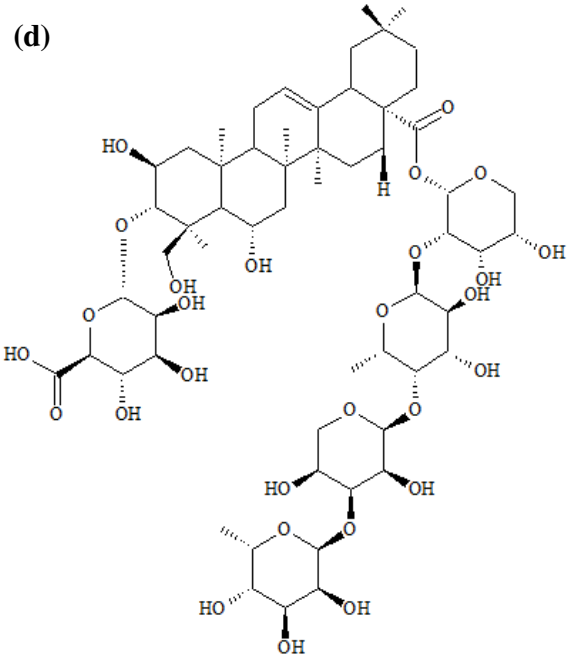
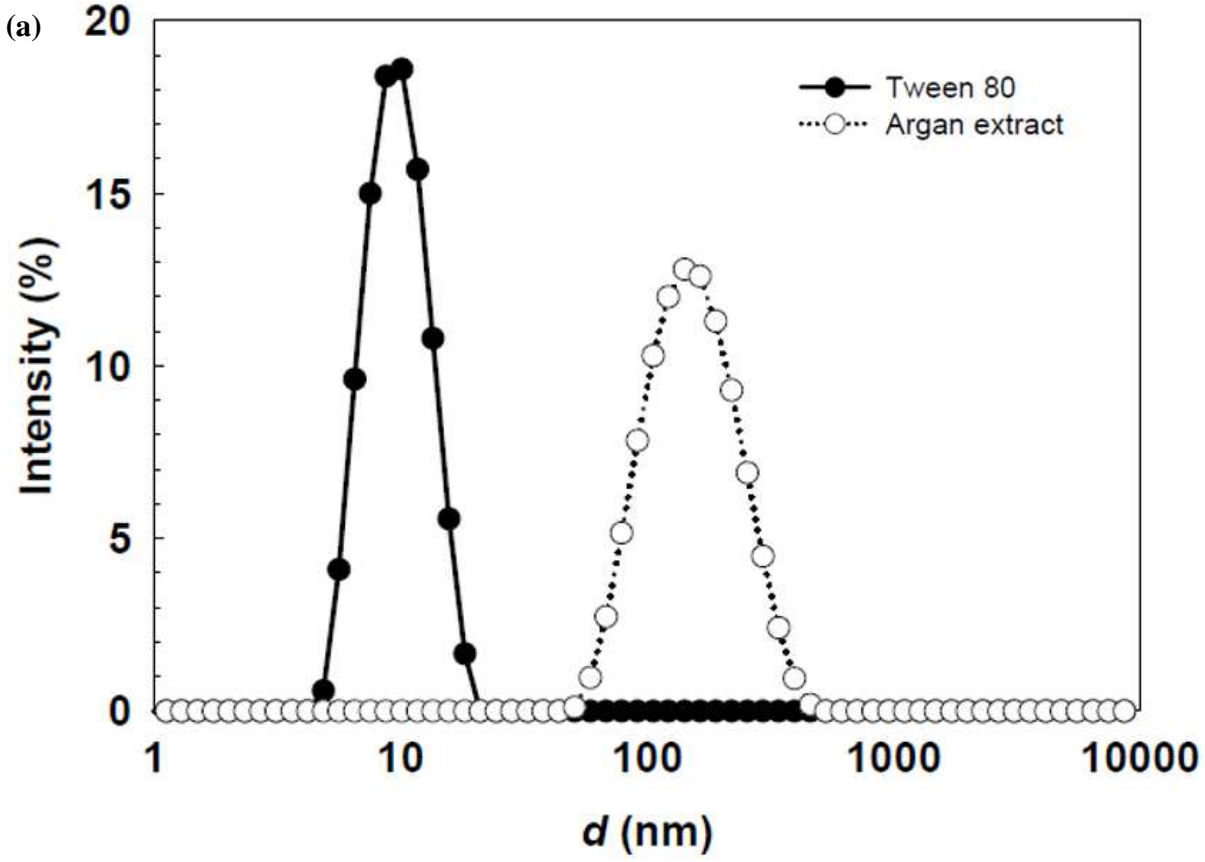


(d)





17 **Figure 3**

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19

20 **Figure 4**

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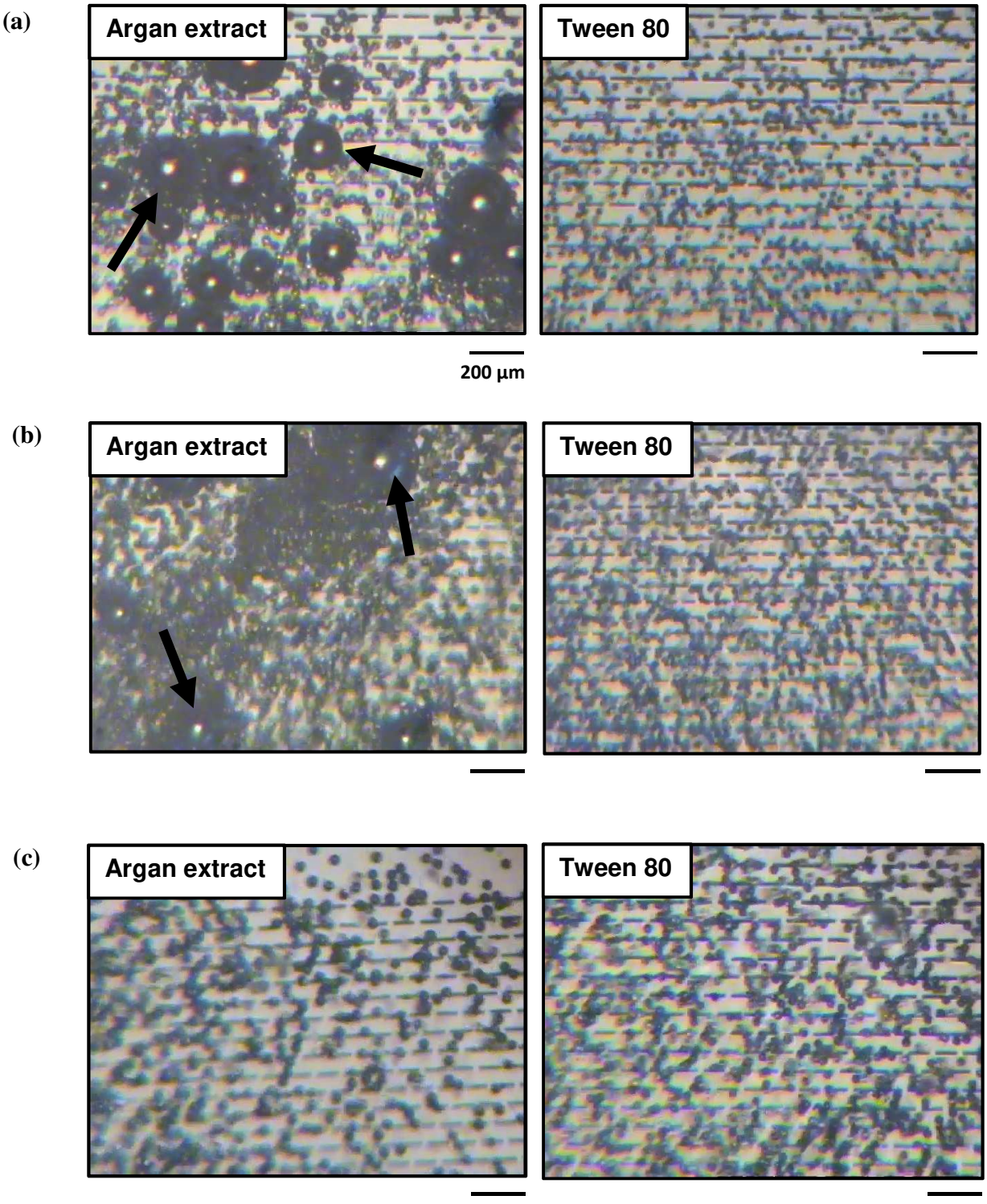
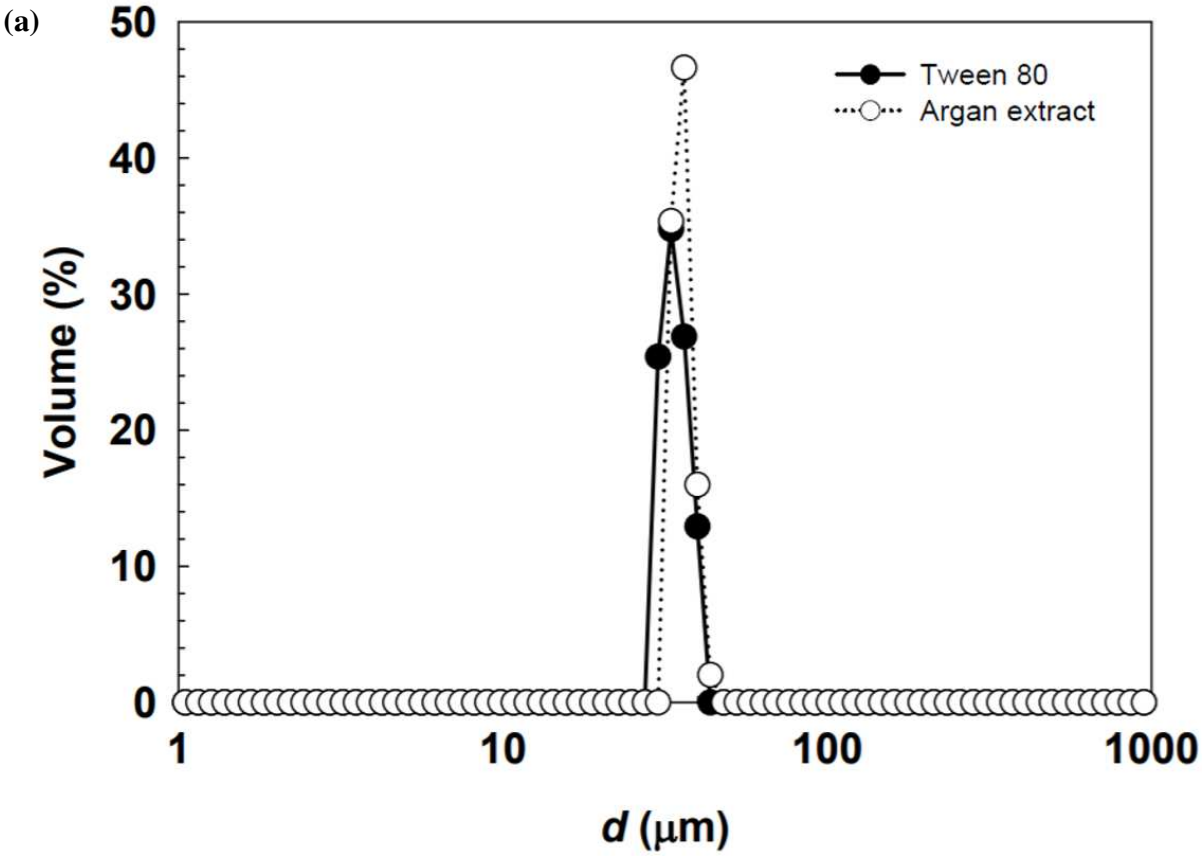
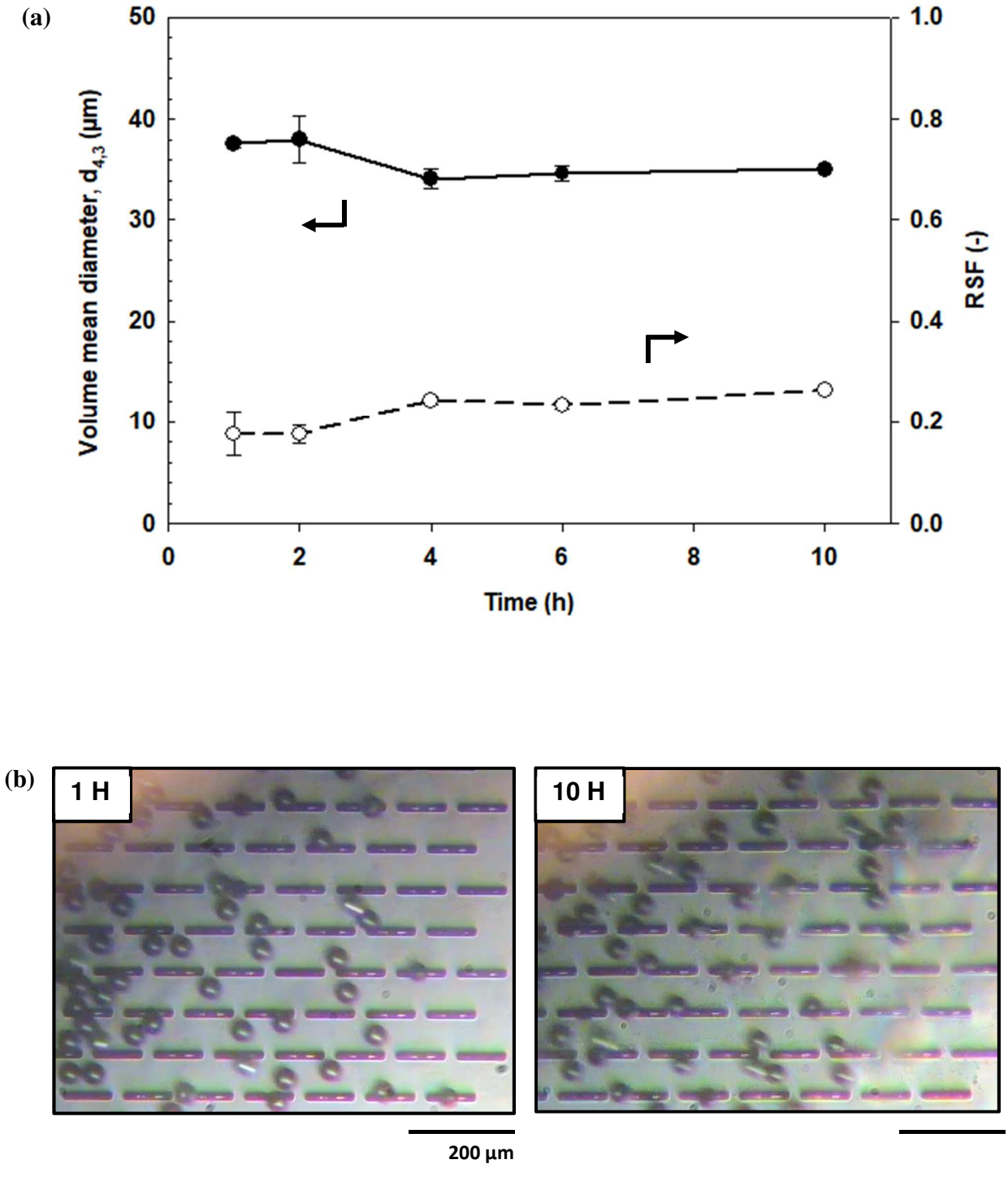


Figure 5

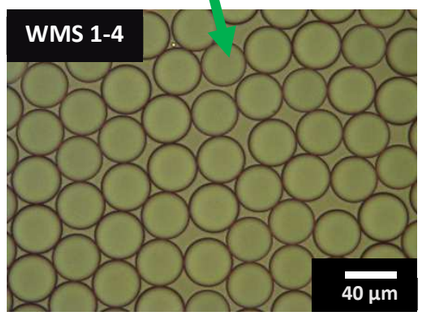

(a)
50
40
30
20
10
0
Volume (%)
1
10
100
1000
d (μm)
Tween 80
Argan extract

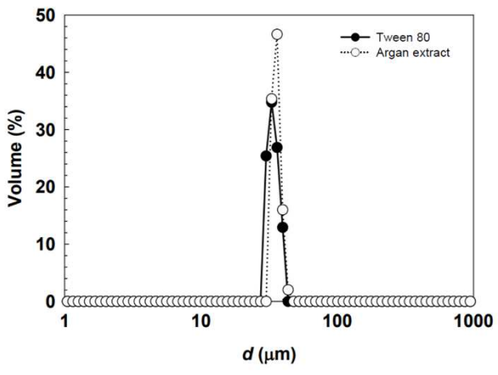
25 **Figure 6**

26



Monodisperse O/W emulsion

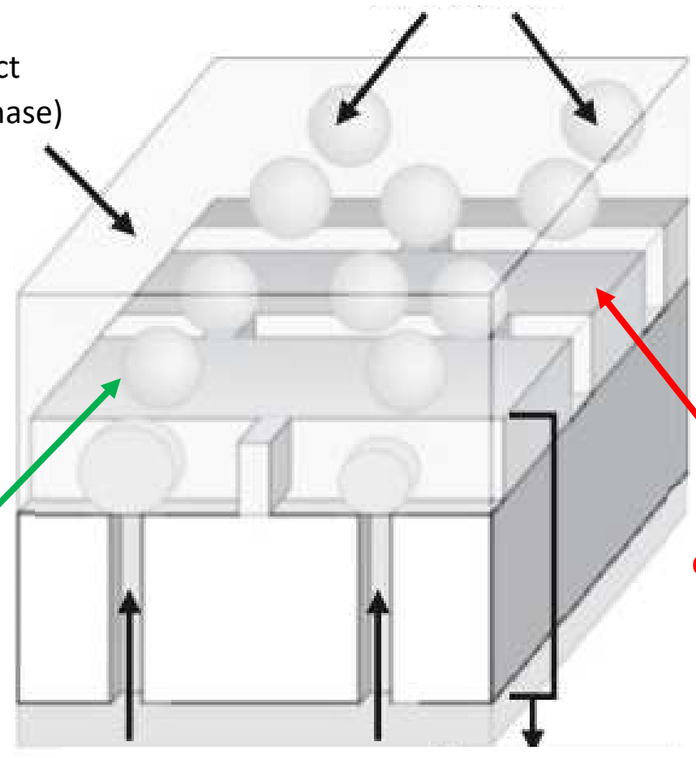




Long MCs (successful MCE)

Argan extract (Continuous phase)

Oil droplets



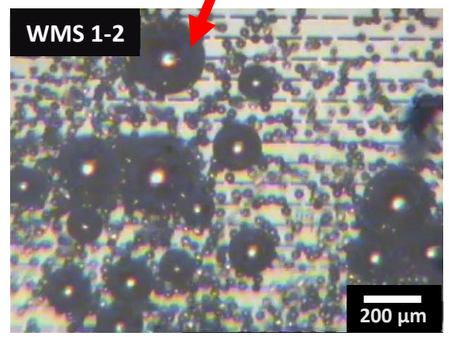
Stabilization of droplets' interface

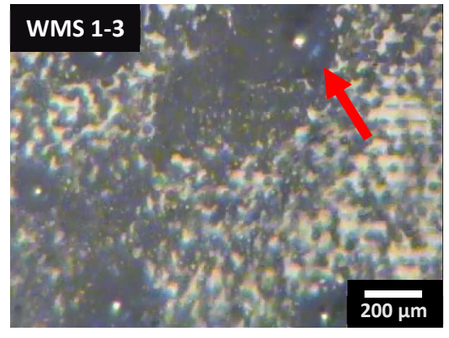
Hydrophobization of MC plate surface

Soybean oil (Dispersed phase)

Microchannels (MCs)

Droplet inflation





Short MCs (unsuccessful MCE)