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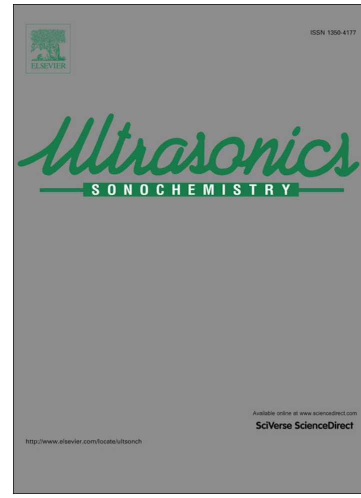

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Revision of the mechanisms behind oil-water (O/W) emulsion preparation by ultrasound and cavitation

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ABSTRACT

Today emulsion preparation is receiving a lot of scientific attention, since emulsions are playing an essential role in many of the big industries, such as food, pharmaceutical or cosmetic industry. One of the most promising techniques for emulsion preparation is ultrasound emulsification. The purpose of this study is to expand the knowledge on the ultrasonically assisted emulsification model, that has not been amended since 1978. The model explains that oil-in-water emulsion formation is a two-step process. Firstly, the surface of the oil phase is disturbed and separated by the acoustic waves. Secondly, cavitation implosions further disrupt and disperse oil drops. We have used a high-speed camera to closely observe oil-in-water emulsion formation. The images show, that the ultrasound emulsification process is profoundly more complex. While the first and the last step of emulsion formation are the same as believed until now, additional intermediate stages of water-in-oil and even oil-in-water-in-oil occur.

Keywords: *ultrasound, emulsion, emulsification, oil-water, cavitation, mechanism, high-speed visualization*

1 INTRODUCTION

Emulsions are heterogeneous mixtures of two immiscible liquid phases, where droplets of one liquid - inner phase, are dispersed into the other - outer or continuous phase [1,2]. Depending which phase is the dispersed one, emulsions are classified as oil in water (O/W), water in oil (W/O) or multiple emulsions (O/W/O or W/O/W), where the dispersed droplets of the inner phase contain even smaller droplets most often of the outer phase [1]. Emulsions are nowadays indispensable and present in numerous different fields such as food industry [6,7], pharmaceutical industry [8,9], cosmetic industry [10] and agriculture [11].

Nowadays, one of the techniques closely investigated for preparation of O/W nano-emulsions is ultrasound emulsification, mainly provoked by cavitation phenomenon [6,8,12]. This is a process where formation, growth and subsequent collapse of bubbles, is a result of high-frequency acoustic irradiation (normally in the range of 20 to 1 MHz) of liquids [14,15,16]. It is believed that the ultrasound emulsification is a two-step process as shown in Fig. 1 [6,8,17–23]. Firstly, interfacial waves, caused by acoustic field, disrupt the stability of the interface between oil and water (Rayleigh-

Taylor instability), causing the separation of the oil phase into the water medium in the form of drops. Secondly, the implosion of cavitation bubbles creates shockwaves, which initiate a chain reaction of a break-up of dispersed drops into smaller droplets until they reach a size where a very stable emulsion is formed [6,8,17–23].

There were also some specific (more application oriented) attempts towards understanding the processes in more detail. It was, for example, found that when a bubble “shuttles« through the water/oil interface emulsification of immiscible phases can occur [24]. Also, the last stages of emulsification process were extensivelly studied from the perspective of influence of the size of the eddies and shear stresses that originate at bubble collapse and can play the dominant role at droplet break-up [25-27].

In the present work, we investigated and recorded the mechanisms behind the formation of an O/W emulsion employing ultrasonication and cavitation phenomenon. To observe these mechanisms more closely, we used a high-speed camera. We firstly investigated the mechanisms on a large (experiments on oil and water in layers) and secondly on a small (experiments on a single oil drop) scale. By observing the occurring mechanisms with a high-speed camera, we were able to determine the mechanisms of the formation of the O/W emulsion in more detail. The emulsion formation proves to be somewhat more complex than thought until now, possibly including also chemical effects caused by cavitation phenomenon. We demonstrated that small oil droplets in water do indeed form in the final stage of the process but in between also O/W and W/O emulsions inside the oil phase are formed.

The new knowledge presented in this paper is a logical step towards a much more detailed study of individual mechanisms. Eventually the novel understanding will enable a more rapid development of emulsion production techniques, which are currently based on pure “trial and error” approach.

2 MATERIAL AND METHODS

2.1 Materials

For all experiments, tap water and sunflower oil were used.

2.2. Experimental set-up

Experiments were performed in the experimental facility at the Department of Power Engineering, Faculty of Mechanical Engineering, University of Ljubljana, Slovenia.

2.2.1 *Ultrasonic bath and piezoelectric transducer*

To study the mechanism taking place in O/W emulsion formation we used a transparent ultrasonic bath already used by Stepišnik Perdih and co-workers [28]. All but one side of the ultrasonic bath were made of acrylic glass. The last side was made of stainless steel. From this side, the ultrasound was emitted at 33 kHz generated by the piezoelectric transducer. The ultrasonic bath was made in a way, that the side emitting ultrasound could be mounted either on the bottom or on the side. This way we could change the direction of the ultrasonic waves. We used the calorimetric method to determine the power of emitted ultrasonic waves [29]. Nominal (full) power of the bath is 100 W. The volume of the bath was 1.1 L.

2.2.2 *Visualization techniques*

Visualization was performed by high-speed camera Photron Fastcam SA-Z. The recording was done at 20.000 fps with resolution 1024×1024 pixels. Shutter time was set to 50 μs. The camera was equipped with Nikon AF-S VR Micro-Nikkor 105mm f/2.8G IF-ED lens mounted together with 6 extension tubes.

Required illumination was provided by the use of two 50.000 lm LED light sources (Ryobi One+). Light sources were facing the camera. This is a common arrangement for cavitation visualization [30]. With such positioning, the cavitation phenomenon and the observed sample are set between the light sources and the camera and therefore appear dark and the background appears bright.

The experimental setup is presented in Fig. 2.

2.2.3 *Microscopic observations*

The 5 mm wide oil drop (after cavitation) was viewed at a 280x magnification using Bresser Biolam microscope.

2.2.4 *Experimental procedure*

We performed two sets of experiments. We started with experiments in layers using larger quantities of oil and water. Observations we made in this set-up led us to perform experiments on a single oil drop submerged in water. In this manner, we were able to study the emulsification process in more detail and drew more firm conclusions.

<u>*Experiments on oil and water in layers*</u>

The ultrasonic bath was carefully filled (to prevent mixing) with water and sunflower oil. The volume ratio of water and oil was 2:1. The ultrasound emitting side was mounted on the bottom of the bath. Hence the propagation of the ultrasound was perpendicular to the water-oil interface. A typical experiment lasted 3 - 4 s, starting with the introduction of the ultrasound. This interval was enough to observe the entire emulsification mechanism. We have obtained the best images when the camera was tilted in a vertical direction for a small angle (approximately 10°). The set up used for these experiments is shown in Fig. 3.

<u>*Experiments on a single oil drop submerged in water*</u>

For these experiments, the oil drop was affixed to the needle tip. For better fixation, we used a wide needle with funnel-shaped tip. The oil drop on the needle tip is shown in Fig. 4. The sample was submerged in the water-filled bath. To minimize the influence of the needle, the ultrasound emitting side was mounted on the side of the bath. We used precise positioning system to place the sample exactly to the pressure antinode. Again, a typical experiment lasted 3 - 4 s.

3 RESULTS AND DISCUSSION

Using the state-of-the-art visualization technique, we gained new insights into emulsification process caused by ultrasonication and demonstrated that the mechanisms behind O/W emulsion formation are not yet fully understood. For the moment our analyses are primarily based on visual interpretation of the phenomenon.

3.1 Results of oil and water in layers experiments

In Fig. 5, two layers, indicated in the figure by dashed lines, are visible. Because the camera was tilted in a vertical direction for a small angle, the oil-water interface is not visible as a line but can be seen as a surface (Fig. 5, Frame b)). Layer on top (brighter) is oil and the bottom layer is water. Because the piezoelectric transducer was mounted on the bottom of the ultrasonic bath the ultrasonic waves propagated from the bottom of the images, toward the top. In the first two frames a) and b) it can clearly be seen that the oil-water interface is disrupted (the interface is not seen as a flat line but rather a curve - marked with dash lines). This is due to the combined effects of ultrasonic waves propagation and cavitation activity. In the Frame c) the inception of a first separating oil droplet is observed at locations where cavitation bubbles appear.

Interesting about newly formed oil droplets is their colour (Fig. 5, Frames d) to j)). The droplets were opaque and not transparent as would be expected if pure oil was torn off. This suggested that the separated droplet content was not homogeneous oil as suggested in numerous published studies from 1978 until today [6,8,17–23] and shown schematically in Fig. 1.

The photographs taken during these experiments (Fig. 5) pointed to the possibility that before the oil droplet is torn off the bulk oil phase due to Rayleigh - Taylor instability (1st Step depicted in Fig. 1), water microjets could be propelled into the bulk oil phase. And the droplet that is torn off the bulk oil phase is rather a W/O emulsion than pure oil (Fig. 5, Frames d) to j)).

Since we wanted to study the observed phenomenon more closely we decided to perform further experiments on a smaller scale – using a single oil drop. By using a single macroscopic oil drop to study the process, we were able to localise the formation of the emulsification process and consequently significantly improve the resolution of the experiments. Also, we could observe all the dynamics seen in Fig. 5 in a single oil drop experiment.

3.2 Results of experiments on a single oil drop

In all the images in this chapter, the ultrasonic transducer was positioned on the side of the bath. Ultrasonic waves, therefore, propagated in the direction from the right, toward the left side, relevant to the images.

To avoid misunderstanding in this chapter, we will use "bulk oil phase" when we refer to single oil drop.

Penetration of water into the bulk oil phase

Opposed to the current understanding of the emulsification mechanism, where clear oil droplets are believed to be separated from the bulk oil phase (Fig. 1, Step 1), we observed that before anything is torn off the bulk oil phase water enters the oil phase (in the present case the macroscopic oil drop).

In Fig. 6 Frame a), we can observe oil-water interface, visible as the curved vertical line. Additionally, the cavitation bubbles are visible on the right side inside the water phase. The following frames (Fig. 6, Frames b) to d)) show, how the cavitation bubbles appear near the interface. When cavitation bubbles implode in the proximity of any boundary, microjets are formed [31]. In the present case, when bubbles collapse near the water-oil interface, water microjet propelled the water through the oil surface into the bulk oil phase. In Fig. 6, Frames c) to j) this is visible as the appearance and growth of a mushroom-shaped structure. In fact, the process seen in the sequence may be a result of several phenomena. The microjet being the predominant one. We base this on visual observation and also on a simple calculation: The bubble size can be estimated from the frequency of the ultrasound. It yields $R=10^{-4}$ m. With the (Rayleigh) collapse time of $t = 10^{-5}$ s and the jet velocity in the order of 100m/s (assuming the bubble is in contact with the interface) we get the approximate length of penetration in an order of a 1mm, which relates well with observations (Fig. 6). On the other hand, the process seems to be much slower than the Rayleigh collapse time, which is, likely a result of acoustic streaming of nuclei towards the interface as a result of primary Bjerknes force [32, 33]. When in the vicinity of the interface the bubbles collapse and push water into the oil.

The cavitation bubbles continuously implode in the vicinity of the O/W interface and, as a consequence, water droplets slowly accumulate near the interface inside the oil.

Emulsion expulsion/tear-off

Only after water penetrated the bulk oil phase, we observed the breakdown of the bulk oil phase into smaller droplets. This is depicted in Figs. 7 and 8. The opaque appearance of the separated droplets implies that it is in fact already an emulsion (as in Fig. 5).

We can see that the small droplet torn off the oil bulk phase is not only oil (due to colour contrast) but rather a W/O emulsion.

After the W/O emulsion droplets are torn off the bulk oil phase they are further exposed to ultrasound waves which causes them to break down into even smaller droplets (Fig. 8, Frame a)). The elongated transparent tube-type shape visible behind the torn off W/O emulsion droplet is an oil trace (Fig. 8, Frames a) and c)). The appearance of cavitation structures and their activity is visible in Frames b) to d). In the Frame e) we can see, that initial droplet is torn off the bulk oil phase (from Frame a)) has been dispersed and the oil trace returned to the oil drop. This process continues until small enough oil droplets are left, which are stable enough to be freely immersed in the continuous water phase and a true O/W emulsion is formed (as proposed in the literature until now).

If we observe the process on a long time and size scales, the breaking off of emulsion droplets from the bulk oil phase can be seen as a fog spreading from the point where O/W emulsion is formed inside the bulk oil phase (Fig. 9).

Further on, the break-up of oil droplets into smaller ones may occur, which is a process governed by the shear forces (Leong et al. [25] for ultrasound and by Vankova et al. [26] and Walstra [27] for single phase turbulent flow]) and is a function of eddy size. This process however, already lies beyond the interest of this work, which deals with the process on how the oil droplets enter the water in the first place.

Formation of an O/W/O emulsion

Sometimes, when the "water into oil" penetration is observed (Fig. 6), the water in oil does not appear homogeneous. It is possible, that not only pure water enters the bulk oil phase, but simultaneously the ingoing water droplets are filled with oil itself leading to an actual O/W/O emulsion formation (Fig. 10).

In Fig. 10, Frames a) to c), a stable droplet inside the bulk oil phase is visible. When this droplet leaves the bulk oil phase (Frames d) to h)), we see that its contents are easily dispersed into the outer water phase. If the separated droplet would only consist of water, the contents should not be visible. Instead, the small "fog" structure is formed (as in Fig. 9), confirming that those are actually small oil droplets, which were entrapped in the water drops inside the bulk oil phase. The formation of an O/W/O emulsion can also be seen in the images taken under the microscope (Fig. 11). The figure shows water droplets in the bulk oil phase. Some water droplets appear clear (marked with letters A to D). However, some of the water drops once again do not appear clear but have non-homogeneous content (marked with numbers 1 to 4). We believe this is the oil inside water droplets inside oil bulk phase suspended on a holder (Fig. 4).

4 CONCLUSIONS

In this paper, we demonstrated that the whole process of formation of O/W emulsion proves to be a lot more complicated than previously thought. What we propose is that the additional steps are amended to the current understanding of the ultrasonic emulsification process. Before the final O/W emulsion is formed, firstly W/O emulsion forms inside the bulk oil phase or even more complex O/W/O. These are later separated from the bulk oil phase and undergo further break down under the influence of ultrasonic waves, and after all these steps are repeated a few times a true O/W emulsion develops. The proposed steps are depicted in Fig. 12.

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REFERENCES

[1] S.R. Reddy, H.S. Fogler, Emulsion stability of acoustically formed emulsions, J. Phys. Chem. 84 (1980) 1570–1575. doi:10.1021/j100449a027.

[2] D.W. A.T. Florence, The formulation and stability of multiple emulsions, Int. J. Pharm. 11 (1982) 277–308.

[3] H. Karbstein, H. Schubert, Developments in the continuous mechanical production of oil-in-water macro-emulsions, Chem. Eng. Process. Process Intensif. 34 (1995) 205–211. doi:10.1016/0255-2701(94)04005-2.

[4] T. Tadros, P. Izquierdo, J. Esquena, C. Solans, Formation and stability of nano-emulsions, Adv. Colloid Interface Sci. 108–109 (2004) 303–318. doi:10.1016/j.cis.2003.10.023.

[5] C. Arancibia, N. Riquelme, R. Zúñiga, S. Matiacevich, Comparing the effectiveness of natural and synthetic emulsifiers on oxidative and physical stability of avocado oil-based nanoemulsions, Innov. Food Sci. Emerg. Technol. 44 (2017) 159–166. doi:10.1016/j.ifset.2017.06.009.

[6] S. Kentish, T.J. Wooster, M. Ashokkumar, S. Balachandran, R. Mawson, L. Simons, The use of ultrasonics for nanoemulsion preparation, Innov. Food Sci. Emerg. Technol. 9 (2008) 170–175. doi:10.1016/j.ifset.2007.07.005.

[7] L. Perugini, G. Cinelli, M. Cofelice, A. Ceglie, F. Lopez, F. Cuomo, Effect of the coexistence of sodium caseinate and Tween 20 as stabilizers of food emulsions at acidic pH, Colloids Surfaces B Biointerfaces. (2018). doi:10.1016/j.colsurfb.2018.02.003.

[8] S.Y. Tang, P. Shridharan, M. Sivakumar, Impact of process parameters in the generation of novel aspirin nanoemulsions - Comparative studies between ultrasound cavitation and microfluidizer, Ultrason. Sonochem. 20 (2013) 485–497. doi:10.1016/j.ultsonch.2012.04.005.

[9] M. Sivakumar, S.Y. Tang, K.W. Tan, Cavitation technology - A greener processing technique for the generation of pharmaceutical nanoemulsions, Ultrason. Sonochem. 21 (2014) 2069–2083. doi:10.1016/j.ultsonch.2014.03.025.

[10] O. Sonneville-Aubrun, J.T. Simonnet, F. L'Alloret, Nanoemulsions: A new vehicle for skincare products, Adv. Colloid Interface Sci. 108–109 (2004) 145–149. doi:10.1016/j.cis.2003.10.026.

[11] L. Zheng, C. Cao, R.Y. Li, L.D. Cao, Z.L. Zhou, M. Li, Q.L. Huang, Preparation and characterization of water-in-oil emulsions of isoprothiolane, Colloids Surfaces A Physicochem. Eng. Asp. 537 (2018) 399–410. doi:10.1016/j.colsurfa.2017.10.031.

[12] K.A. Ramisetty, A.B. Pandit, P.R. Gogate, Chemical Engineering and Processing : Process Intensi fi cation Ultrasound assisted preparation of emulsion of coconut oil in water : Understanding the effect of operating parameters and comparison of reactor designs, Chem. Eng. Process. Process Intensif. 88 (2015) 70–77. doi:10.1016/j.cep.2014.12.006.

[13] D.J. McClements, S.M. Jafari, Improving emulsion formation, stability and performance using mixed emulsifiers: A review, Adv. Colloid Interface Sci. 251 (2018) 55–79. doi:10.1016/j.cis.2017.12.001.

[14] M. Klavarioti, D. Mantzavinos, D. Kassinos, Removal of residual pharmaceuticals from aqueous systems by advanced oxidation processes, Environ. Int. 35 (2009) 402–417. doi:10.1016/j.envint.2008.07.009.

[15] P.C. Sangave, A.B. Pandit, Ultrasound pre-treatment for enhanced biodegradability of the distillery wastewater, Ultrason. Sonochem. 11 (2004) 197–203. doi:10.1016/j.ultsonch.2004.01.026.

[16] I. Tzanakis, G.S.B. Lebon, D.G. Eskin, K.A. Pericleous, Characterizing the cavitation development and acoustic spectrum in various liquids, Ultrason. Sonochem. 34 (2017) 651-662.

[17] M.K. Li, H.S. Fogler, Acoustic emulsification. Part 1. The instability of the oil-water interface to form the initial droplets, J. Fluid Mech. 88 (1978) 499–511. doi:10.1017/S0022112078002232.

[18] M.K. Li, H.S. Fogler, Acoustic emulsification. Part 2. Breakup of the large primary oil droplets in a water medium, J. Fluid Mech. 88 (1978) 513–528. doi:10.1017/S0022112078002244.

[19] K.S. Suslick, The Chemical Effects of Ultrasound, Sci. Am. 260 (1989) 80–86. doi:10.1038/scientificamerican0289-80.

[20] J.P. Canselier, H. Delmas, A.M. Wilhelm, B. Abismaïl, Ultrasound Emulsification—An Overview, J. Dispers. Sci. Technol. 23 (2002) 333–349. doi:10.1080/01932690208984209.

[21] A. Cucheval, R.C.Y. Chow, A study on the emulsification of oil by power ultrasound, Ultrason. Sonochem. 15 (2008) 916–920. doi:10.1016/j.ultsonch.2008.02.004.

[22] M. Kaci, S. Meziani, E. Arab-Tehrany, G. Gillet, I. Desjardins-Lavisse, S. Desobry, Emulsification by high frequency ultrasound using piezoelectric transducer: Formation and stability of emulsifier free emulsion, Ultrason. Sonochem. 21 (2014) 1010–1017. doi:10.1016/j.ultsonch.2013.11.006.

[23] L. Patil, P.R. Gogate, Ultrasound assisted synthesis of stable oil in milk emulsion: Study of operating parameters and scale-up aspects, Ultrason. Sonochem. 40 (2018) 135–146. doi:10.1016/j.ultsonch.2017.07.001.

[23] F. Seniha Güner, Y. Yağci, A. Tuncer Erciyes, Polymers from triglyceride oils, Prog. Polym. Sci. 31 (2006) 633–670. doi:10.1016/j.progpolymsci.2006.07.001.

[24] S. Zhao, Z. Dong, C. Yao, Z. Wen, G. Chen, Q. Yuan, Liquid–Liquid Two-Phase Flow in Ultrasonic Microreactors: Cavitation, Emulsification, and Mass Transfer Enhancement, American Institute of Chemical Engineers vol. 64 (2018)

[25] T.S. H. Leong, T.J. Wooster, S.E. Kentish, M. Ashokkumar. Minimising Oil Droplet Size using Ultrasonic Emulsification, Ultrason Sonochem. 2009 Aug;16(6):721-7.

[26] N. Vankova, S. Tcholakova, N.D. Denkov, I.B. Ivanov, V.D. Vulchev, and T. Danner, Emulsification in turbulent flow - 1. Mean and maximum drop diameters in inertial and viscous regimes, J. Colloid Inter. Sci 312 (2) (2007) 363-380.

[27] P. Walstra. Principles of emulsion formation. Chemical Engineering Science. Volume 48, Issue 2, January 1993, Pages 333-349

[28] T. Stepišnik Perdih, B. Širok, M. Dular, On the bubble-surfactant interaction, Chem. Eng. Process. Process Intensif. 121 (2017) 198–204. doi:10.1016/j.cep.2017.08.016.

[29] T.A. T. Kimura, T. Sakamoto, J.-M. Leveque, H. Sohmiya, M. Fujita, S. Ikeda, Standardization of ultrasonic power for sonochemical reaction, Ultrason. Sonochem. 3 (1996) S157–S161. doi:10.1016/S1350-4177(96)00021-1.

[30] I. Biluš, B. Bizjan, L. Lešnik, B. Širok, B. Pečnik, M. Dular, Non-contact method for analysis of cavitating flows, Ultrasonics. 81 (2017) 178–186. doi:10.1016/j.ultras.2017.03.011.

[31] T.J. Mason, Ultrasound in synthetic organic chemistry, Chem. Soc. Rev. 26 (1997) 443–451. doi:10.1039/CS9972600443.

[32] A. A. Doinikov, Bjerknes forces and translational bubble dynamics Bubble and Particle Dynamics in Acoustic Fields: Modern Trends and Applications, (2005).

[33] I. Tzanakis, G.S.B. Lebon, D.G. Eskin, K.A. Pericleous, Characterizing the cavitation development and acoustic spectrum in various liquids, Ultrasonics sonochemistry 34, (2017), 651-662.

Figure Captions

Figure 1. Current understanding of the O/W emulsion formation (adopted from [17]).

Figure 2. Scheme of the ultrasonic bath and piezoelectric transducer.

Figure 3. Set-up for experiments on oil and water in layers.

Figure 4. Oil drop suspended on a holder.

Figure 5. Separation of a W/O emulsion drop from the bulk oil phase.

Figure 6. Collapse of the cavitation bubble forms a water jet, which penetrates the oil-water interface and pushes water into the oil bulk phase.

Figure 7. Break down of the bulk oil phase surface stability and separation of a W/O emulsion droplet.

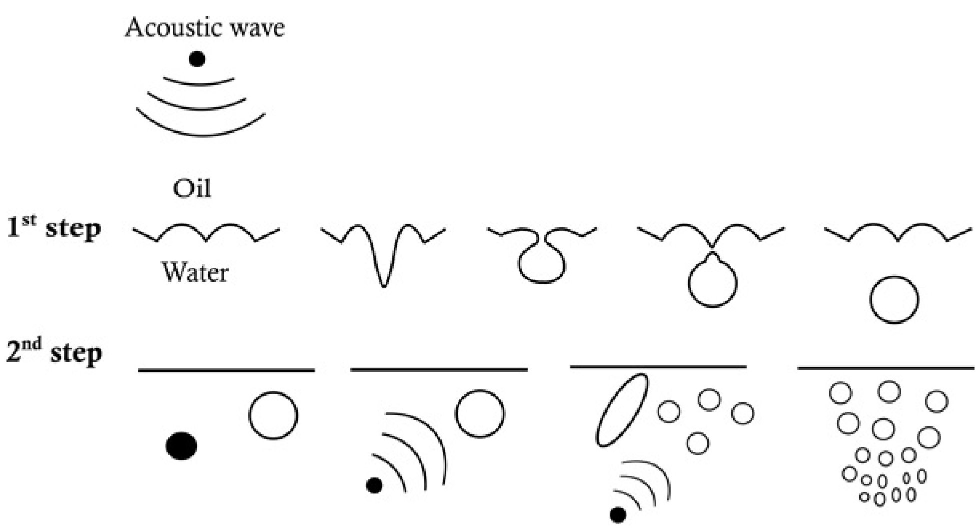
Figure 8. Further breakdown of a W/O emulsion.

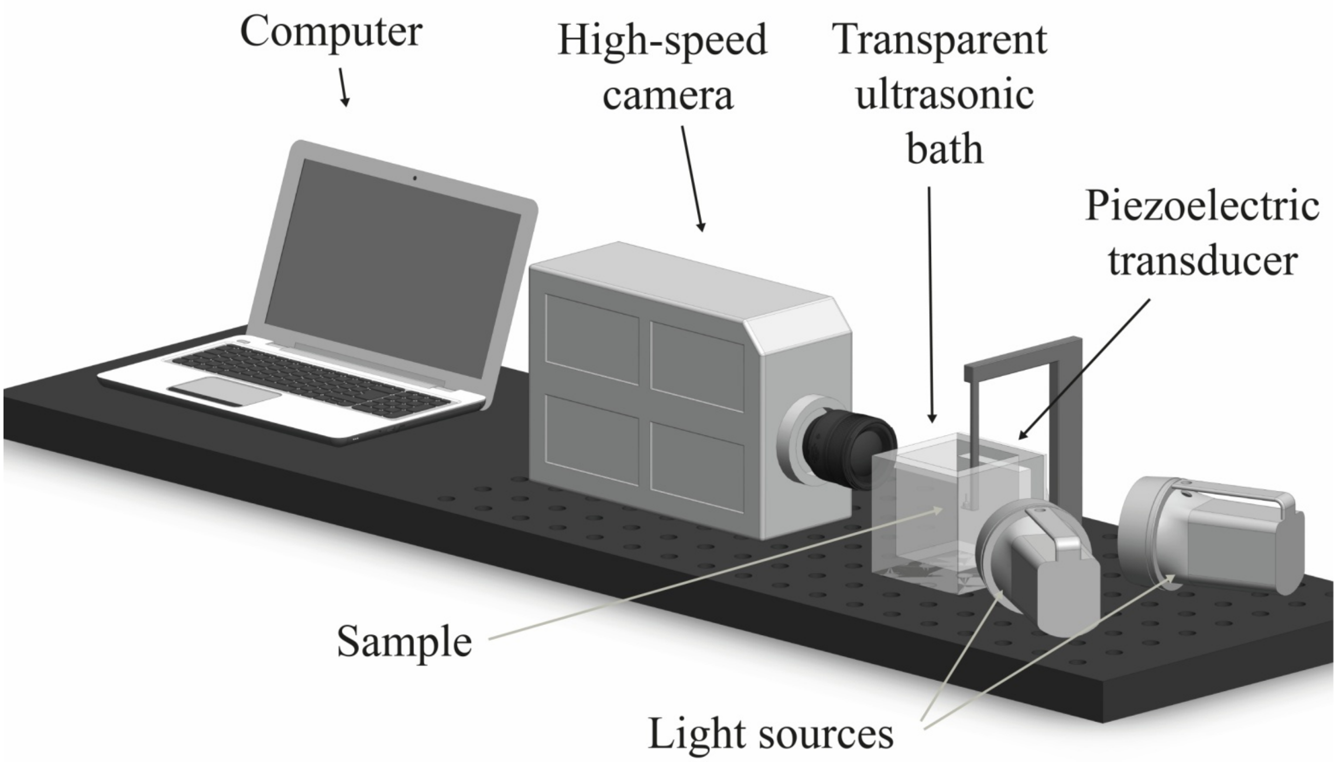
Figure 9. Fog formation around the oil bulk phase.

Figure 10. O/W emulsion formation inside the oil bulk phase and its break down outside the oil bulk phase.

Figure 11. Microscopic images of O/W emulsion droplets inside a 5 mm oil phase after cavitation.

Figure 12. Proposed new model of O/W emulsion formation.





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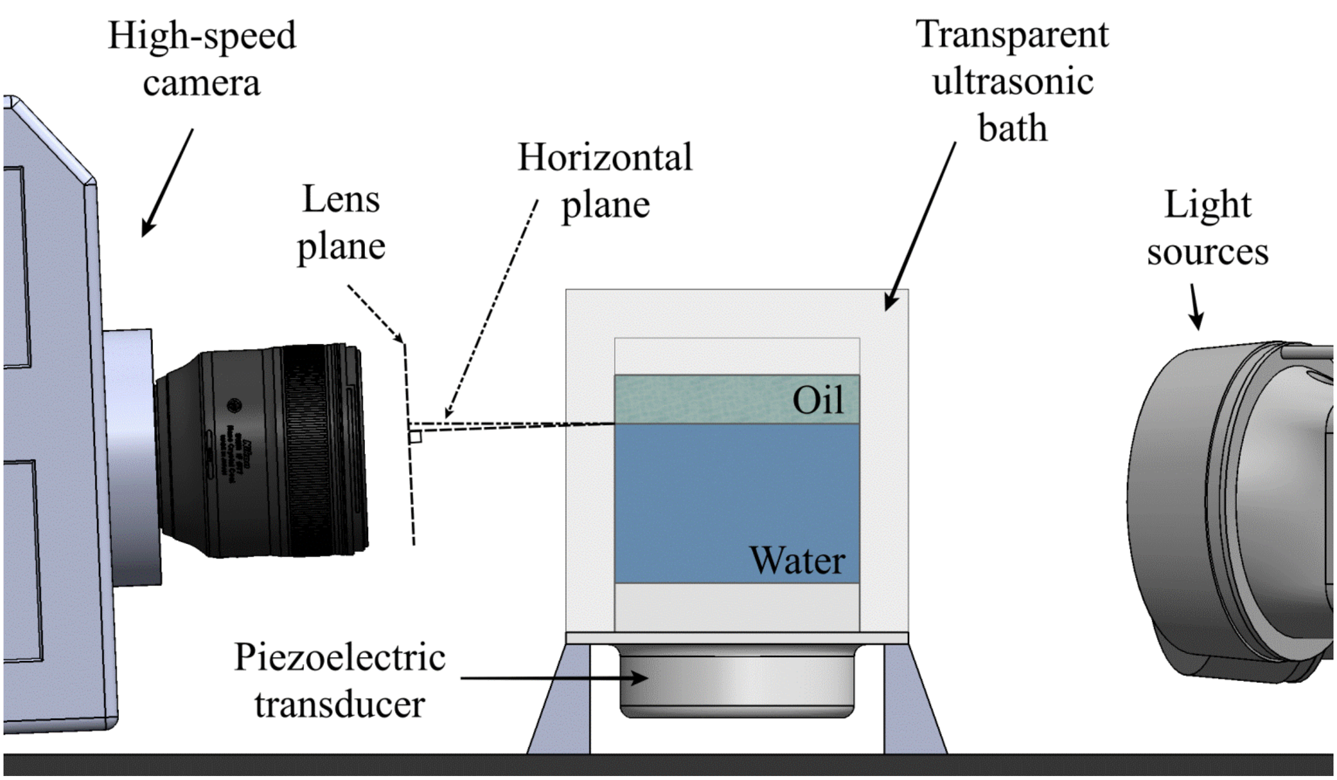
Piezoelectric transducer
Sample
Light sources



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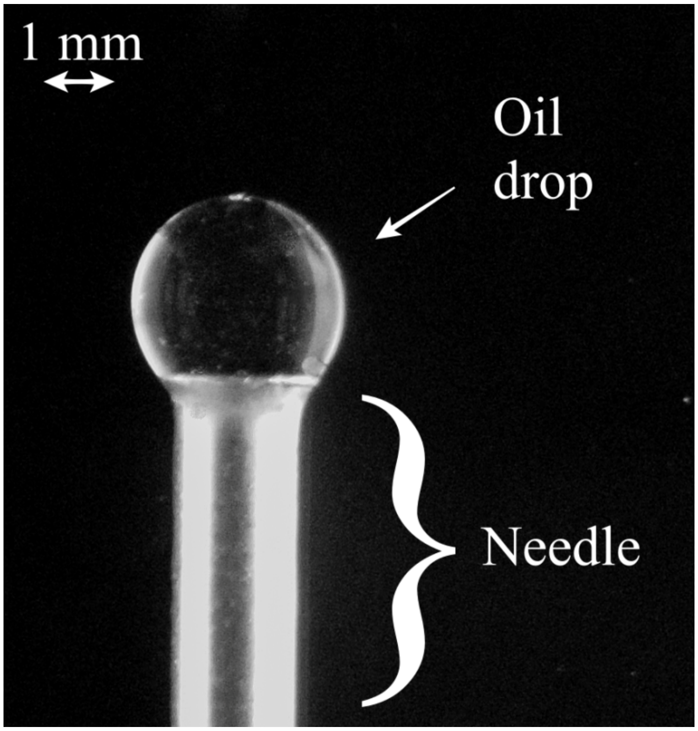
Transparent ultrasonic bath
Horizontal plane
Lens plane
Light sources
Oil
Water
Piezoelectric transducer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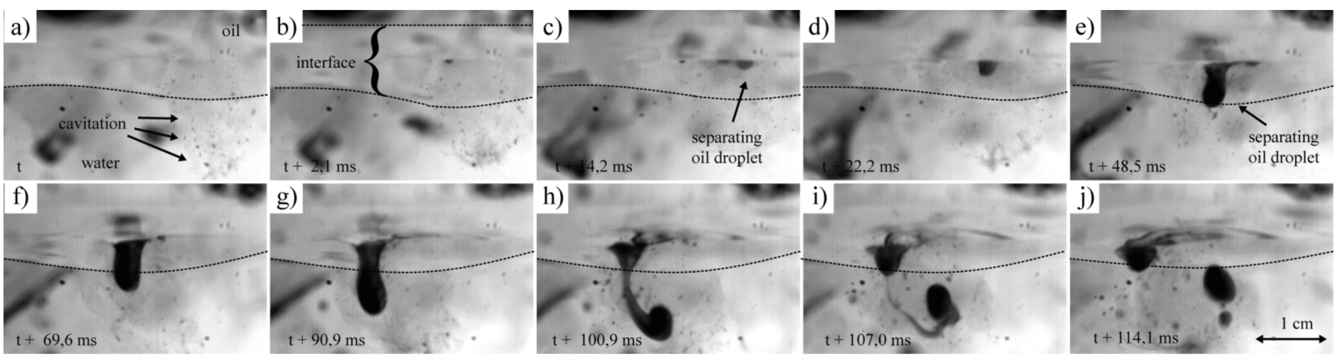
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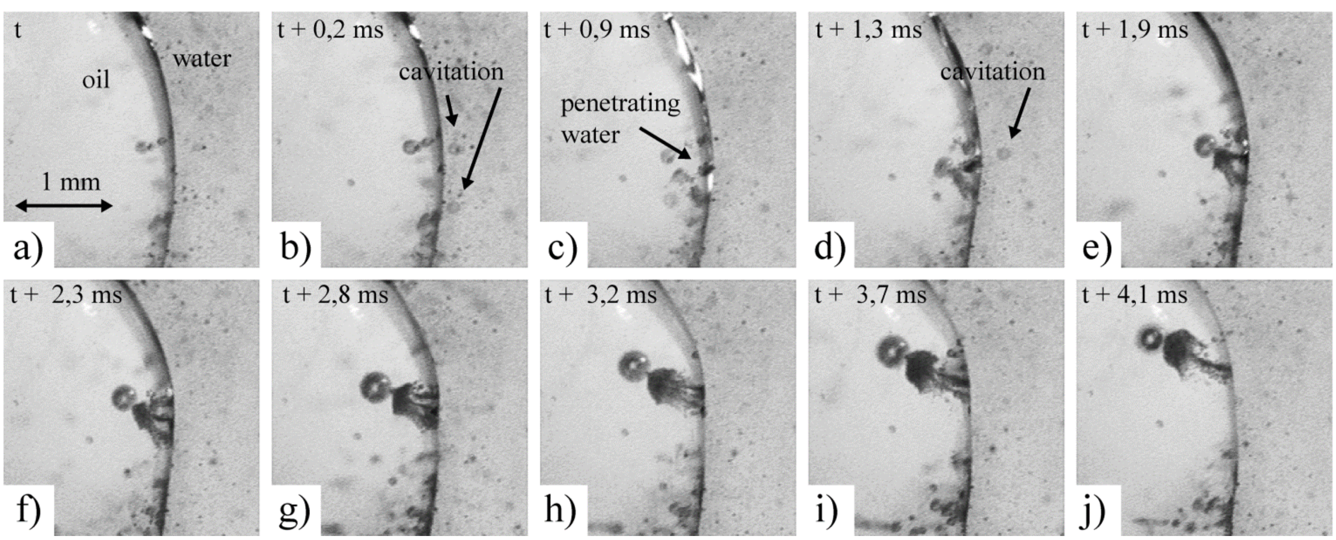
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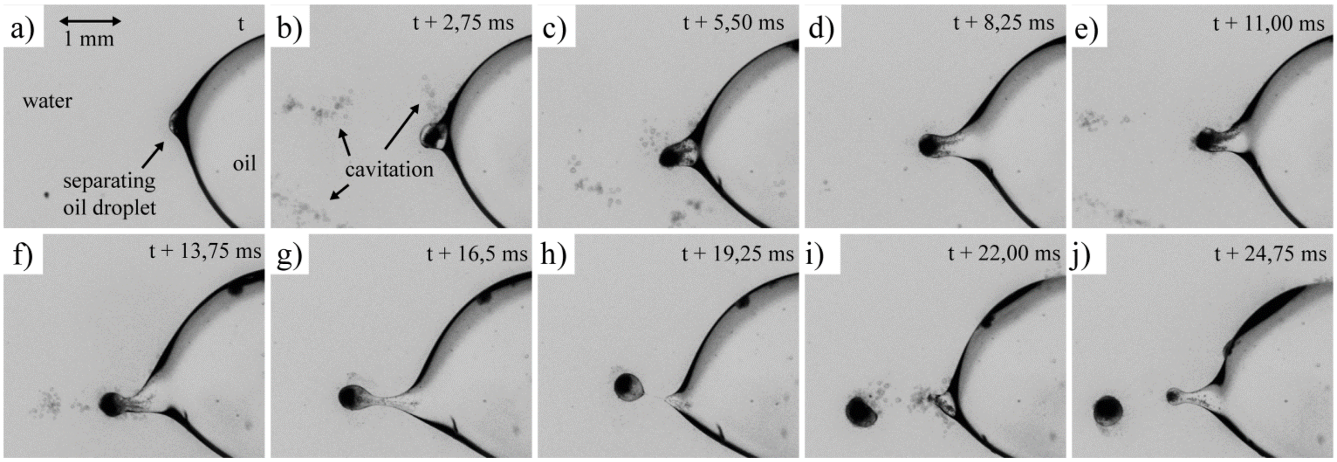
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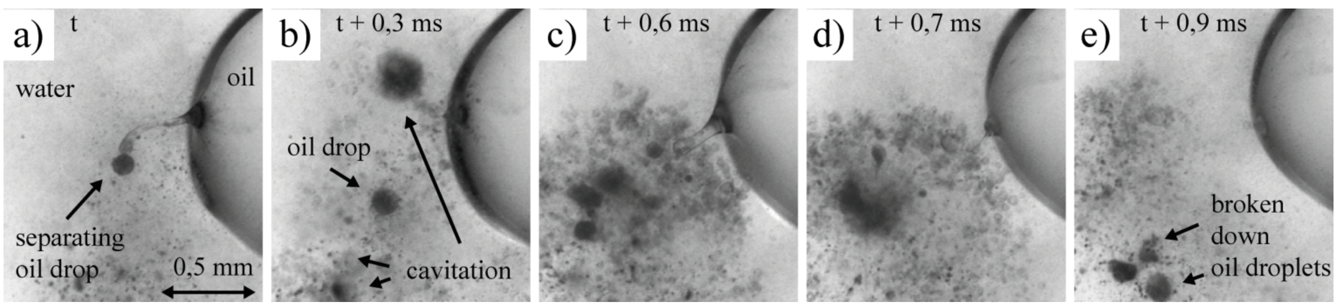
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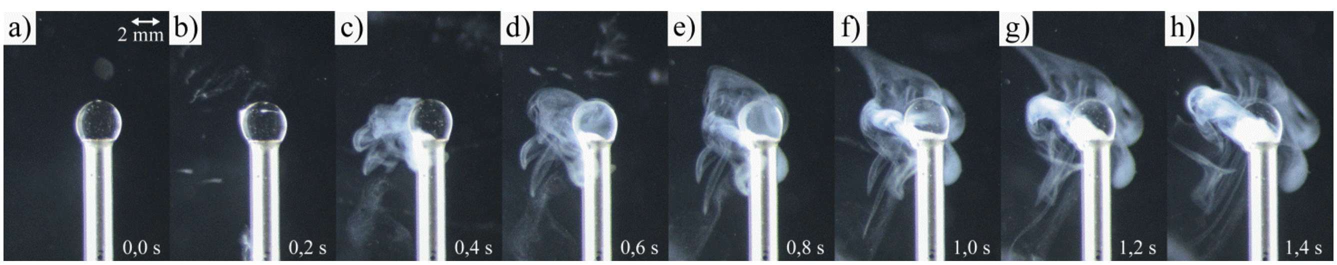
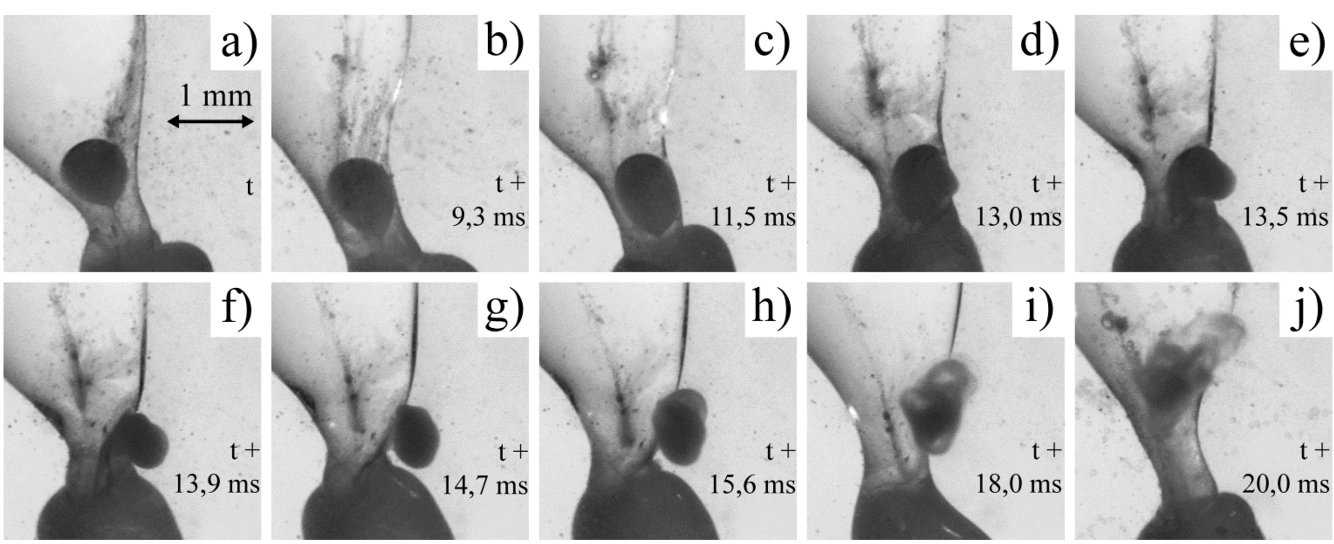
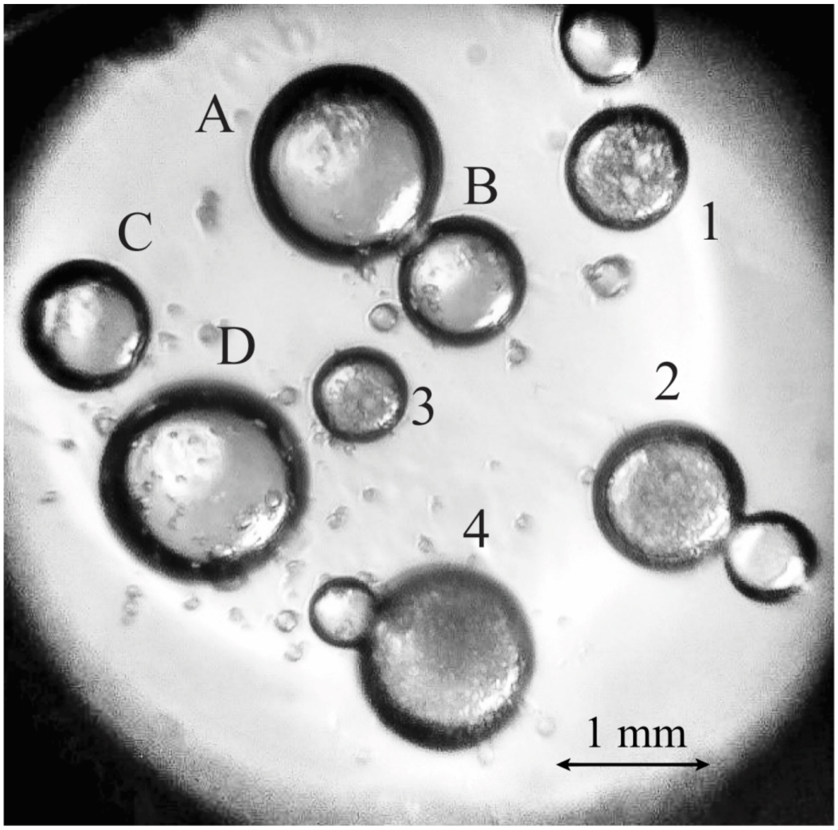
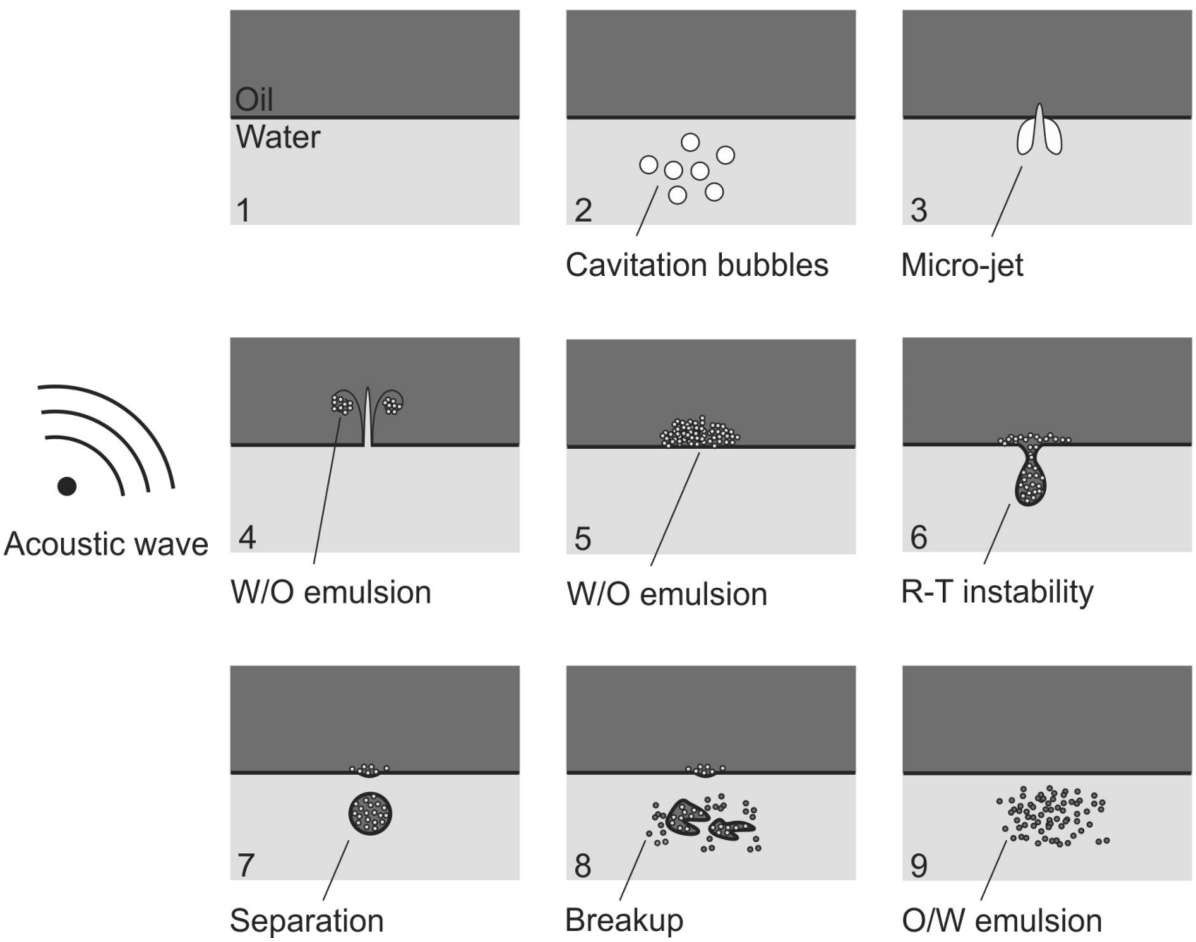
t
water
oil
separating
oil drop
0,5 mm
b)
t + 0,3 ms
oil drop
cavitation
c)
t + 0,6 ms
d)
t + 0,7 ms
e)
t + 0,9 ms
broken
down
oil droplets


a)
2 mm
0,0 s
b)
0,2 s
c)
0,4 s
d)
0,6 s
e)
0,8 s
f)
1,0 s
g)
1,2 s
h)
1,4 s


a)
1 mm
t
b)
t +
9,3 ms
c)
t +
11,5 ms
d)
t +
13,0 ms
e)
t +
13,5 ms
f)
t +
13,9 ms
g)
t +
14,7 ms
h)
t +
15,6 ms
i)
t +
18,0 ms
j)
t +
20,0 ms


A
B
C
D
1
2
3
4
1 mm


Oil
Water
1
2
Cavitation bubbles
3
Micro-jet
Acoustic wave
4
W/O emulsion
5
W/O emulsion
6
R-T instability
7
Separation
8
Breakup
9
O/W emulsion

HIGHLIGHTS

- O/W emulsion preparation by ultrasound is more complex than believed until now
- High-speed camera recordings reveal additional intermediate W/O and O/W/O stages
- Mechanical effects driving ultrasonic emulsification are discussed
- The new ultrasonic emulsification model is proposed