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Emulsification by ultrasound : Relation between intensity and emulsion quality

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Ultrasonic emulsification of oil and water was carried out and the effect of position of the ultrasound source from the interface on emulsion quality was studied using ultrasonic bath and horn. Correlations for the effect of distance of the ultrasound source from the interface on various emulsion properties such as dispersed phase fraction, droplet diameter were developed. Large variation in the emulsion properties with small changes in the position of ultrasound source was observed and correlated with an exponential type of equation. Discrepancies in the results of heterogeneous liquid phase systems reported in the literature were attributed to the small changes in the location of ultrasound source. Severe attenuation of ultrasound intensity by the oil layer was quantitatively established using decomposition of aqueous KI solution as a model reaction. The droplet diameter was predicted using Kolmogorov eddy length model. The collapse pressure developed in cavitation was determined indirectly and compares favorably with the reported values. The highly localised nature of cavitation phenomenon is also well established as a cause for the variation in the emulsion quality.

Heterogeneous liquid-liquid systems hold an important place in the domain of chemical engineering. The success of the above systems that may be reacting or non-reacting, depends on the ability to generate high interfacial area by an equipment economically. In precise, formation of a fine emulsion is a prerequisite. Many a times, one has to resort to techniques such as Phase Transfer Catalysis(PTC), to overcome the reaction rate limitation due to low interfacial area. These catalysts are not only costly, but also quite hazardous. Ultrasound assisted cavitation generating equipment could be a better solution especially when fine emulsions are required. The quality of emulsion generated by sonication is far more superior to that of conventional processes. The most important advantage is that the generation of an emulsion do not require addition of surfactants.

Cavitation is the formation (when surrounding pressure falls below vapour pressure), growth, and violent collapse of the microbubbles/cavities. Numerous ways are available for generation of cavitation[1]. Ultrasound as a source is the one that has been studied extensively and commercially exploited to a significant extent. Two types of cavitation occur based on the life of the cavity. When the cavity life is less than one acoustic cycle, it is called transient cavitation. The other type that does not satisfy the above criteria is stable cavitation. Cavitational intensity is maximum when the cavitation is transient. The driving frequency (usually 25 kHz) of the ultrasonic source should be less than the natural frequency of the cavity for the cavitation to be transient[2]. For water and most other liquids, the above conditions are satisfied easily.

Cavitation is a way to concentrate the energy of the ultrasound locally, but at numerous places simultaneously. Positioning of the ultrasonic source is a vital parameter affecting the quality of emulsion. Extensive study of this parameter has been done here. This resulted in understanding some of the fundamental aspects of cavitation phenomenon also.

Emulisfication phenomenon

Emulsification under the conditions of

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ultrasonic irradiation occurs in two stages. In the beginning, disturbances are generated at the interface due to the vibrations as a result of the passage of ultrasound and this gives rise to fairly coarse drops due to the phenomenon of Rayleigh-Taylor instability mechanism[3,4]. For this stage, the velocity needed to rupture the large interface need not be high. The typical velocity generated in the liquid on the passage of the sound (calculated indirectly) is of the order of 0.1 m/s and this is sufficient to rupture large interfaces. Further breakage of fine droplets requires high inertial forces to overcome the surface tension forces ($2\sigma/R$) which increase as the size of the droplets decrease. Cavitation plays an important role at this stage. Near the liquid-liquid interface, the velocity of the wall of the imploding cavity could reach as high as 150 m/s due to the imbalance in the rush of liquid[5]. This process is called acoustic streaming. The turbulence created by this process, along with the shock waves of the collapse, may tear off part of the droplet when present near the collapsing cavity. Thus continuous breakage of the droplets occurs in the cavitating zone up to a critical size that is the characteristic of the particular system[3]. The maximum size of the droplet can be predicted with the Kolmogorov eddy length theory[2] and is given by,

$$d_{\max}=C\varepsilon^{-2/5}\gamma^{3/5}\rho_c^{-1/5} \quad \ldots (1)$$

where C is a constant and usually of the order of unity, ε is the average power dissipation per unit volume, γ and ρ_c are interfacial tension and density of the continuous phase respectively. The above equation is used frequently, though it can predict only the order of magnitude of the maximum drop diameter.

Experimental Procedure

Equipment—The source of ultrasonic energy for emulsification was an ultrasonic horn of driving frequency 22.7 kHz, rated power input of 240 W and an ultrasonic bath of driving frequency 22 kHz, rated power input of 120 W. The emulsion was analysed using IPPLUS image analyser, by preparing and observing slides under microscope.

Emulsification using ultrasonic horn—To a 100 mL beaker, 30 mL of distilled water and 40 mL of edible oil were added as shown in Fig. 1. The sample was sonicated for 15 min after positioning the horn at 3 mm from the oil-water interface, in the upper oil layer above. At the end of sonication, two emulsions were obtained. The upper layer is the water dispersed in oil (Upper emulsion) and the lower layer is the oil dispersed in water (Lower emulsion). The temperatures of both the emulsions were measured at the end of sonication. The densities of both the emulsions were determined inorder to calculate the weight fraction of the dispersed phase in two emulsions. Droplet size of the dispersed phase in both the emulsions was analysed using an image analyser attached to a microscope. This method directly gives the maximum, minimum and average size of the droplets in the sample. Similar experiments were performed by increasing the distance of horn from the interface to 5 mm and 7 mm.

Decomposition of aqueous KI solution—The study was conducted to quantify the intensity of

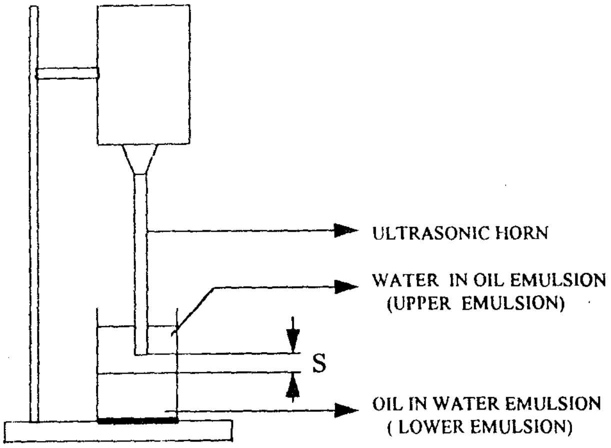

Fig. 1—Emulisification with Ultrasonic Horn

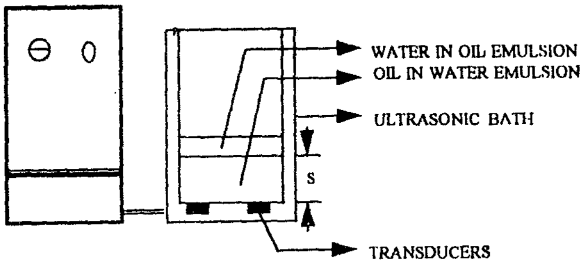

Fig. 2—Emulisification with Ultrasonic Bath

Equipment	Distance (mm) m	Density-Upper Emulsion (g/mL)	Density-Lower Emulsion (g/mL)	% Water In Upper Emulsion	% Oil In Lower Emulsion	Average Drop Dia. Upper Emulsion (Microns)	Average Drop Dia. Lower Emulsion Microns	Interfacial area-upper emulsion (m^2/m^3) $*10^{-2}$	Interfacial area- lower emulsion (m^2/m^3) $*10^{-2}$
HORN	3	0.9252	1.0021	3.40	2.32	1.89	1.99	1709.4	699.5
	5	0.9225	1.0030	0.12	1.22	3.14	2.51	22.9	291.6
	7	0.92245	1.0035	0.06	0.61	4.13	3.31	8.7	110.6
BATH	3	0.9255	0.9837	3.65	24.87	2.49	2.20	879.52	6782:7
	5	0.9229	0.9847	0.61	23.65	2.21	1.53	165.61	9274.0
	7	0.9225	0.9865	0.02	21.44	1.39	1.18	8.63	10901.7

ultrasound reaching the interface and the extent of cavitation occurring in the aqueous phase. Sound intensity reaching the aqueous phase generates cavities, which collapse subsequently resulting in high local temperatures and pressures. This results in the cleavage of the water molecules present inside the cavity to OH$^\bullet$ radicals, which combines to form H_2O_2 molecules. The H_2O_2 molecules oxidise the KI present in the solution, resulting in the liberation of iodine which can spectrophotometrically be measured. One percent solution of KI was prepared using distilled water and this was used as aqueous phase along with few drops of starch solution for emulsification instead of distilled water. The clear aqueous phase in the lower emulsion was obtained by breaking the emulsion and the absorbance of the aqueous phase was measured using UV/VIS Spectrophotometer at 355nm. The I_2 concentration in lower emulsion could thus be estimated. The above procedure was repeated for various distances of the horn from the interface.

Emulsification using ultrasonic bath—The experimental procedure for the bath is similar to that for the ultrasonic horn (Fig. 2). Here the distance between the ultrasound source (present beneath the bottom surface of the bath) and liquid-liquid interface was varied using different volumes of water. Also the volume of the oil used was 100 mL due to larger cross-sectional area of the bath. The sonication period was 15 min. The temperature and density of both the emulsions were measured. The emulsions were analysed using image analyser attached to a microscope.

Results and Discussion

The intensity of ultrasound decreases with an increase in distance between the ultrasonic source and liquid- liquid interface[6]. This is mainly due to the attenuation of sound by the molecules of the liquid. In the presence of dispersed phase, the attenuation is more severe[7]. The decrease varies exponentially with distance and is given by the equation of the form[5,6],

$$I=I_o\exp(-2\alpha S) \qquad \ldots (2)$$

where I_o is the intensity at the tip of the horn, S is the distance from the source of ultrasound and α is the attenuation coefficient. The attenuation coefficient is directly proportional to the viscosity of the liquid. Hence position of the ultrasound source is an important parameter in any acoustic emulsification process.

Table 1 summarises the effect of position of the ultrasonic source on the emulsion quality, defined in terms of phase fractions and droplet sizes. The dispersed phase fraction decreases with increase in distance (S) from the interface for both ultrasonic horn and bath. The droplet diameter increases with increase in distance for horn. The trend for the bath is altogether opposite. The detailed analysis on the effect of position of ultrasonic source on each property is discussed below.

Mechanical efficiency of the ultrasonic equipment—For emulsification with a horn, temperatures of both the emulsions were measured after sonication. The temperature difference between the upper and lower emulsion was found to be 12 K which is due to the difference in the specific heat capacities and volume of active cavitating zone of the two emulsions. The effect of position of the ultrasonic source from the interface had a marginal effect on the pattern of temperature difference. Considering the temperature rise in both emulsions, the energy input was calculated by calorimetric method[5] and the efficiency of the ultrasonic equipment was determined as:

$$\eta_{\text{mechanical}} = \frac{\Sigma m_{li} c_{pli} (\Delta t)_l + \Sigma m_{ui} c_{pu\,i} (\Delta t)_u}{\text{energy input}} \times 100 \quad \ldots (3)$$

Where m is the mass of the liquid, c_p is the specific heat capacity of the liquid and (Δt) is the rise in temperature. The subscript 'l' refers to the lower emulsion, 'u' refers to the upper emulsion and 'i' represents the various phases present in the emulsion (i.e., oil and water). The efficiency of the horn and bath were found to be 2% and 14% respectively. The higher energy efficiency of the bath over the horn is primarily due to the fact that energy is delivered to liquid over a larger cross-sectional area. The standard configuration of the horn supports more accessories such as cooling fan for transducer. Moreover, the bath is thoroughly insulated. These factors may also contribute for the higher energy efficiency of the bath. This clearly indicates the scope for increasing the mechanical performance of the ultrasound equipment and thereby making the process more energy efficient and economical.

Dispersed phase hold-up—In both the upper and lower emulsions, dispersed phase fraction decreases with increase in the distance of the horn from the interface. The generation of coarse droplets, which is the first stage of emulsification, is mainly dependent on the inertial forces generated by the vibration of liquid at the interface due to the passage of ultrasound. As the interface moves away from the horn, the oscillatory motion of the liquid near the interface reduces due to the reduced intensity of sound. Also, the process of acoustic streaming that is necessary for the formation of a liquid jet at the interface for the transfer of one liquid phase into another, decreases due to reduced cavitational activity at the interface. This reduction is again caused by the attenuation of sound by the oil due to its high viscosity. For the horn, the dependence on distance is given by the following correlation based on the experimental observations.

% oil in lower emulsion (F_{OL})
$$= 6.317{*}\exp(-0.334\ S)\ (R^2 = 0.999) \quad \ldots (4)$$

% water in upper emulsion (F_{WU})
$$= 621.66{*}\exp(-4.813\ S)(R^2 = 0.957) \quad \ldots (5)$$

It can be found that the reduction is more severe with respect to the upper emulsion than to the lower emulsion. As already mentioned, the oil severely attenuates the passage of sound on account of its higher viscosity. At larger distances of the horn from the interface, very little energy reaches the water layer. Thus the overall cavitational activity and resultant acoustic streaming are reduced in the lower emulsion. Hence the transfer of water into oil layer is more severely affected by the distance (S) as compared to the transfer of oil into the lower emulsion.

In case of a bath, the dispersed phase fraction in the upper layer is higher than the lower layer by near an order of magnitude. This can be attributed to the fact that thickness of the oil layer is comparatively smaller than the water layer below because of the large cross-sectional area of the bath and the lesser quantity of oil taken for emulsification (100 mL). Moreover, as a large area of the liquid is coupled to the bath surface that is vibrating, the total momentum transferred to the liquid is higher. Both factors create a good mixing of the upper layer, resulting in an increased presence of water in the upper emulsion. The dispersed phase fraction in the lower emulsion is comparable with that of ultrasonic horn.

The effect of the position of the interface from the bottom surface of bath follows the same trend as with the ultrasonic horn. It decreases exponentially as the distance S increases. The relation based on the experimental observation is,

% oil in lower emulsion(F_{OL})
$$= 30.248{*}\exp(-0.169\ S)\ (R^2 = 0.845) \quad \ldots (6)$$

% water in upper emulsion(F_{WU})
$$= 41.118{*}\exp\ (-0.836\ S)\ (R^2 = 0.836) \quad \ldots (7)$$

Similar to the horn, the rate of decrease of the dispersed phase with an increase in distance is higher with respect to the upper emulsion than the lower emulsion.

Effect on droplet size—The droplet diameter of dispersed phase in both the emulsions increases with increased distance of the ultrasonic horn from the interface. The breakage of coarse droplets, which is the second stage of emulsification, requires high inertial forces. This occurs mainly due to turbulence generated by the collapsing cavities present near the droplet (acoustic streaming). When ultrasound intensity near the interface varies, the process of breakage of coarse droplets, which are generated continuously near the liquid-liquid interface, is affected due to reduced cavitational activity in terms of number as well as intensity of cavity collapse. The dependence of drop diameter on distance S for horn based on experimental results is,

$$d_{\text{water}} = 1.093 * \exp(0.195\ S)\ (R^2 = 0.937) \quad \ldots (8)$$

$$d_{\text{oil}} = 1.349 * \exp(0.127\ S)(R^2 = 0.998) \quad \ldots (9)$$

In the case of ultrasonic bath this dependence is given by,

$$d_{\text{water}} = 4.083 * \exp(-0.156\ S)(R^2 = 0.897) \quad \ldots (10)$$

$$d_{\text{oil}} = 3.450 * \exp(-0.146\ S)(R^2 = 0.991) \quad \ldots (11)$$

The ultrasound intensity near the interface decreases as we move the tip of the horn away from the interface, resulting in a reduced cavitational intensity and an increase in droplet diameter. The dependence of ultrasound intensity on the position of source is opposite to that in the ultrasonic bath. For a height of liquid in the bath less than half the wavelength of sound, intensity of ultrasound increases with an increase in height of the liquid level[8]. Since cavitational collapse intensity and the resultant acoustic streaming are directly proportional to ultrasound intensity, the droplet diameter decreases with an increase in height of the liquid. Though drop diameter increases with S, the dispersed phase holdup decreases with an increase in S, indicating that the acoustic streaming is not the governing phenomenon involved in the transfer of one phase into another. As mentioned above, formation of coarse droplets require very little inertial forces near the interface and this occurs mainly due to the momentum transferred by the vibrating surface. Thus the inertial force near the interface decreases as the interface moves away from the vibrating surface. For the horn, the area of vibrating surface coupled to the liquid is very low and the momentum transferred to liquid is also correspondingly lower. Thus the acoustic streaming near the interface is the dominant phenomenon in the horn for the transfer of one phase into another, which depends on the ultrasound intensity near the interface. Hence the dispersed phase holdup decreases with increase in distance S.

At higher liquid levels, standing waves are generated, resulting in an nonhomogenous intensity distribution and prediction of the performance of the bath becomes difficult. This phenomenon stands against using a bath in large scale systems. Also droplet diameter in the upper emulsion is higher by 18- 45 percent for both horn and bath, irrespective of the position of ultrasound source. This indicates that the cavitational activity in the upper emulsion responsible for breakage is less intense. The cavitation threshold of oil is higher due to its high viscosity and low vapour pressure and viscous forces acting on the cavity dampens its violent collapse, resulting in reduced cavitational activity. The mean droplet diameter is known to be proportional to the viscosity of continuous phase to the power 0.1. i.e., $d_p \alpha (\mu_{\text{oil}}/\mu_{\text{water}})^{0.1}$. The oil used in the study had a viscosity of about 35 cp. This suggests an increase in the droplet in the droplet diameter in the oil layer by a factor of 1.41. The observed increase is of similar magnitude.

Effect on interfacial area—Interfacial area of emulsion depends on both dispersed phase fraction and droplet diameter. The maximum value of interfacial area obtained in this study for horn is 1.71 E05 m^2/m^3 and 6.98 E04 m^2/m^3 for upper and lower emulsions respectively when the position of the horn is near the interface. Similarly, for the bath, the maximum value for lower emulsion (8.79 E04 m^2/m^3) occurs when the position of liquid-liquid interface is closer to the bottom surface of bath. For other distances refer to Table 1. The value for the upper emulsion is very high due to a very large dispersed phase fraction. For all cases, the interfacial area decreases drastically with increase in the distance of ultrasound source from the liquid-liquid interface. Hence for reactions that are masstransfer controlled, movement of ultrasonic source away from the interface will severely affect the reaction rate.

Decomposition of potassium iodide—As already mentioned, the objective of this reaction is to support the fact that a decrease in emulsion quality when the position of the horn is away from the interface is due to a reduced sound intensity reaching the interface and aqueous layer. Decomposition of aqueous KI solution is a model reaction commonly used to quantify cavitational intensity, by measuring the liberation of iodine[9]. These results then can be used to represent the extent of attenuation of sound by oil layer qualitatively. Fig. (3) shows the concentration of iodine present in the aqueous layer with respect to distance.

Liberation of I_2 depends upon the generation of $OH^\bullet$ radicals by the cleavage of water molecule, which in turn depends upon the cavitational intensity. Thus it can be a good indicator of the intensity of sound reaching the lower emulsion and hence of the cavitational intensity in that phase. Like all other properties of the emulsion, there is a decrease in liberation of iodine with increase in distance of the horn away from the interface. The iodine concentration exponentially decreases with increase in distance.

$$I_2 \text{ Liberation(I2)} = 3.069E\text{-}05 * \exp(-0.462\, S)(R^2 = 0.999) \qquad \ldots (12)$$

The value of I_2 liberated, when the horn was dipped just below the interface, in the aqueous KI solution, matches with the previous reported value[10]. Fig. 4 gives a comparative picture of properties of the emulsion, as well as the results of KI decomposition in which the ordinate is dimensionless. The subscript 'max' represents the maximum value of the variable that is considered in the ordinate and this occurs either at 3 mm or 7 mm. The identical trend of the curves for iodine liberation as well as average droplet diameter indicates that these variables are solely dependent on the cavitation intensity, as predicted earlier. This trend also compares favorably with experimental cavitation intensity measurements[6].

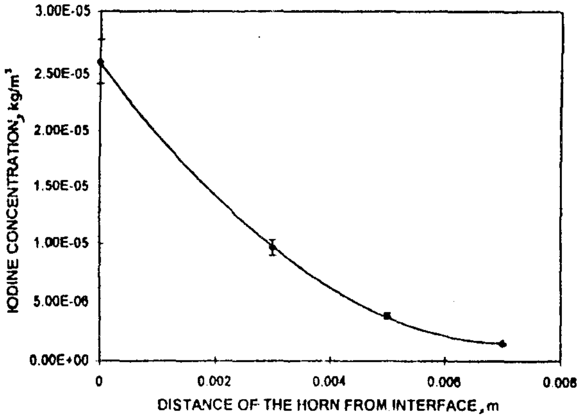

Fig. 3—Effect of Position of the Horn on Iodine Liberation

Prediction of droplet size and collapse pressure of cavity—As can observed from Fig. (3), the amount of iodine liberated becomes negligible when the distance is 0.007 m. Thus almost all the energy must be getting dissipated within this distance. Considering that the total energy (numerator of Eq. (3)) is dissipated in a hemispherical zone of radius 0.007 m below the horn tip, the d_{max} in the upper emulsion can be predicted using Eq. (1). This was found to be 23 μm, which is higher than the observed maximum droplet diameter of 7.6 μm. This indicates that the energy dissipation per unit volume should be far higher than that used for calculation at least locally due to cavitation. Rearranging Eq. (1), the energy dissipation per unit volume required to break a droplet of average diameter of 7.6 μm can be computed from the following equation,

$$\varepsilon = \left(\frac{\gamma^{\frac{3}{5}} \rho_c^{-\frac{1}{5}}}{d_{\max}} \right)^{2.5} \qquad \ldots (13)$$

For oil in water emulsion, this value comes out to be 1.0319*E09 W/m^3 which is higher than that can be dissipated in conventional mixing systems. The experimental value based on criteria of uniform energy dissipation within the zone considered is 2.5510*E06 W/m^3. Thus the actual energy dissipation must be higher by three orders of magnitude to obtain a droplet of size measured or the value of energy is dissipated in only 0.25% of the total volume of the zone considered. In ultrasonic cavitation, it is the second phenomenon that is taking place. This compares favorably with

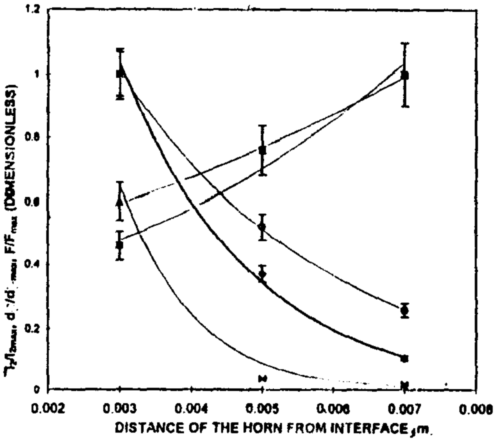

Fig. 4—Effect of Position of the Horn on Emulsion Properties ◆—Iodine liberation, ■—Avg. drop dia.-Upper emulsioin, ▲—Avg. drop dia.-Lower emulsion, ×—Dispersed phase fracton-Lower emulsion, O—Dispersed phase fraction-Upper emulsion.

the experimental cavity volume fraction (0.3%) where the total number of cavities were measured indirectly[7]. Thus the mechanism of energy dissipation in cavitation is highly localised .

The approximate pressure developed during cavity collapse can also be calculated indirectly. The velocity of the jet of liquid necessary for the droplet breakage can be determined by assuming the critical Weber number for droplet breakage to be equal to 1.

$$W_b = \frac{\Delta\rho(\Delta u)^2 r}{\gamma} \quad \ldots (14)$$

where r is the radius of the droplet and Δ represent the difference in properties between dispersed and continuous phase. The lowest diameter of the droplet detected in the emulsion was 1 μm. As the droplet is practically stationary, Δu effectively denotes the velocity of the continuous phase (liquid jet) surrounding the drop. Assuming that the pressure energy during the cavity collapse has been completely converted into kinetic energy of the liquid jet that breaks the drop, the collapse pressure generated can be calculated as,

$$P = \frac{1}{2}\rho_c(\Delta u)^2 \quad \ldots (15)$$

For a droplet diameter of 1 μm, the value is nearly 24 atm. So the pressure developed during the cavity collapse should be greater than this value. This value of collapse pressure compares favorably with the experimental measurement of 36 atm as the maximum collapse pressure in the same system[6].

Conclusion

Ultrasound has good potential as a source of energy for emulsification. It can be advantageously used in liquid-liquid heterogeneous systems especially when the reactions are controlled by mass transfer. Ultrasonic bath has inherent limitations due to generation of standing waves and low intensity of ultrasound. A horn can be effectively used for the emulsification. The dependence of the emulsion properties studied on position of the ultrasound source can be generalised by the following equation.

$$\text{Emulsion property} = A \exp(B.\,S)$$

where A and B are constants. All the processes (droplet breakage and decomposition of KI) which depend solely on cavitation show identical trends with respect to position of ultrasonic horn (Fig. 4). Positioning of the horn near to the liquid-liquid interface has yielded favorable results. Also the prediction of decrease in emulsion quality due to decreased intensity of the sound reaching the interface for cavitation is well confirmed by the aqueous KI decomposition reaction. Thus this reaction can also be used to estimate the magnitude of attenuation of the sound intensity for any liquid immiscible with aqueous KI solution. The rapid changes in the properties of the liquid- liquid heterogeneous system with minor changes in the position of ultrasound source could also be an important reason for the poor reproducibility of the results of numerous reactions reported in literature.

Acknowledgement

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Nomenclature

d —drop diameter, µm
d_{water} —drop diameter of water in upper emulsion, µm
d_{oil} —drop diameter of oil in lower emulsion, µm
F —dispersed phase holdup (%)
I —ultrasound intensity, W/m^2

I_o —ultrasound intensity near the source, W/m^2
S —distance of the interface from the ultrasound source, m
u —velocity of liquid, m/sec
Greek
ε —average energy dissipation, W/m^3
η —efficiency of ultrasound equipment
α —sound attenuation co-efficient
σ —surface of the liquid, N/m
μ —viscosity of the liquid, kg/ m sec
γ —interfacial tension, N/m
ρ —density of the liquid, kg/m^3
Subscipts
c —continuous phase
OL —oil in water/ lower emulsion
WO —water in oil/ upper emulsion

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