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Comparison CFD modeling of drop-interface coalescence and experimental results

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ABSTRACT

In the present work, separation of two immiscible liquids (sodium hydroxide and dimethyl disulfide) was investigated and coalescence of dimethyl disulfide with mother phase was studied at the interface by using numerical simulations. This phenomenon is a part of Merox unit by gravitational section in it. The volume of the fluid method has been used for modeling which droplet of the dispersed phase is released from nozzle toward the interface. Several experiments for measurement of coalescence time of the droplets of the dispersed phase (Dimethyl Disulfide) with the mother phase, using a high speed camera (fps1000), were performed. The comparison between experimental results and the results of modeling were compared and its shown that a compatibility between them and also the effect of several parameters including of drop diameter, continuous phase density regulated by temperature, and sodium hydroxide concentration, continuous phase to dispersed phase viscosity ratio on coalescence time were investigated.

Keywords*: coalescence time; two immiscible liquid; Independent variable; Interfacial tension; CFD modeling.*

1. INTRODUCTION

Separation of two immiscible liquids is on the most important phenomenon that be used in oil and gas industry . One of the processes used for LPG sweetening process is the Merox liquid-liquid process[1].

In this process, mercaptans react with alkaline solution (sodium hydroxide) to sodium mercaptides in an extractor and then mercaptides are oxidized to disulfides in an oxidizer. The combination alkaline solution with disulfides enters the gravity separator. Separation of these two immiscible liquids is significantly important since they provide the reutilization of caustic and limit the entrance of disulfide into the extraction cycle. The separation of the phases is mostly governed through the coalescence of the droplets and the mother phase at the interface (drop-interface coalescence), it is convenient to have a better understanding of this phenomenon. The first article about coalescence between drop and interface was investigated by Charles and Mason in 1960 [2].They studied the residence time , rupture of the film and the effect of some parameters such as density, surface tension and viscosity. During the 1980s and 1990s taking advanced numerical techniques to simulate the interaction was done by most of the researchers [3]. Uemura et al. Investigated different stages of interfacial phenomenon. They observed that when a droplet passes through the interface between two immiscible liquids, the coalescence process will involve four consecutive stages [4]. In the first stage, the droplet rising through the interface pulls up the heavier liquid and collision of liquid bodies will happen .heavier liquid around it. In the second stage, this film of the heavier liquid ruptures at the front of the droplet. In the third stage drainage of the thin film is occurred and in the last stage merging of the liquids is done completely. There are some mathematical model and numerical simulation of drop coalescence and passages of drop through a quiescent liquids interface, Manga and Stone studied the effect of physical properties on the evaluation of interface based on the numerical model [5]. A numerical model based on Navier Stockes equations for passage of a single droplet through the interface was reported by Shopov and Minev [6]. CFD (Computational fluid dynamic) simulation based on VOF of the passages of droplet through the interface was investigated by Bonhomme et al. They used different densities, viscosities and droplet size over a wide range on the simulation [7]. Gebauer etal. investigated single droplet coalescence of toluene water by CFD modeling. They also reported a detailed description of local phenomena, like film rupture, velocity gradients, pressures and micro-droplet entrainment [8]. Wang et al. studied the dynamic behavior of the coalescence between two droplets with different temperatures in oil system with using the volume of fluid (VOF) method[9].

Eulerian-Lagrangian simulation of bubble coalescence wasc studied by xue et al. They investigated the bubble collisions and the contact time with this modeling[10]. CFD model was validated in a bubble column with organic liquids by Guo et al, The result of modeling waown a good predictions because it quantitatively described the effect of liquid properties on the bubble size, interphase forces, turbulence parameters, and bubble breakup and coalescence behaviors[11]. politova et al. studied the effects of drop size, surfactant chain-length and concentration and viscosity of the oil phase on the stability of water drops, towards planar oil-water interface. They found that The small drops coalesce before the formation of a planar film with the large interface The results was compatible by theoretical models[12].

Because of the lack of advancement in imaging technology in the 1980s, the limited amount of work on coalescence and passages of droplet through the interface was studied. The beginning of the new century the digital imaging technology was progressed and it contributed to a series of excellent works on drop coalescence [13].

In this study we employed a high speed camera (fps 1000) to capture the coalescence steps and passages of droplet through the interface and also CFD simulation using VOF method were also carried out with two immiscible liquids and passages of a dispersed droplet through the interface. The comparison between results was carried out. By carefully varying a parameter while keeping the rest constant, one can improve the operating condition in this type of process and provide a good foundation for the design of many industries such as separation, deemulsification and environmental examination

2. SETUP OF EXPERIMENT MATERIAL

Due to the effect of various operational parameters and chemical phenomena on the coalescing on the interface between two phases, a cylindrical glass device with an inner diameter of 8cm and height of 40cm, designed and it was made by the workshop. This arrangement is intended to inject three different angles. A digital camera with a high frame rate (fps1000) was used in extracting images. In this experiment, a direction and imaging were considered and camera frame rate was 500 fps up to 1000 fps. To create high quality images in this test, a resolution 1920 x 720 pixels was considered. The camera is placed to indicate movement of drops and it focused on the interface. Set up of the experiment is shown in Fig. 1.

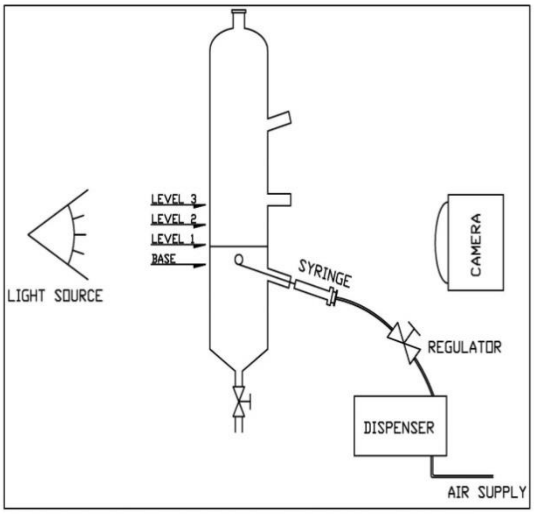

Figure 1. Setup of the Experiment.

2.1 Materials.

The list of chemicals used in this study is Sodium Hydroxide flakes, Dimethyl Disulfide, Demineralized water for making solution. All chemicals have over 99 percent purity. For increasing accuracy and repeatability of tests, Merck chemicals have been used.

2.2 Computational approach and Computational Domain, Boundary, and Initial Conditions.

For modeling of gravity separator in above experiment, we used the commercial software ANSYS Fluent. The mass and momentum conservation equations solved are given by eq1 and eq 2. The fluids are considered incompressible.

$$\nabla \vec{v}=0 \qquad (1)$$

$$\partial(\rho \vec{v})/\partial t+\nabla.(\rho \vec{v}\,\vec{v})=-\nabla p+\nabla\tau+\rho g+F \qquad (2)$$

where $\vec{v}$ is the fluid velocity vector, ρ and μ are the effective fluid density and viscosity, p is the static pressure, τ is the viscous stress tensor, $\vec{g}$ is acceleration due to gravity, and $\vec{F}$ is the volumetric body force due to surface or interfacial tension. By using this equations the modeling is defined. The typical computational domain used in the simulation is shown in Figure 2. The liquids in the simulation considered are Newtonian. In this simulation is assumed that dispersed drop raise up in a straight line on continuous phase. For preventing wall effects on drop coalescence a 2D domain with dimension 10*10 cm is chosen on simulation. The middle of the computational domain is introduced as the initial position of liquid– liquid interface.and drop is released on three level in the heavy phase.

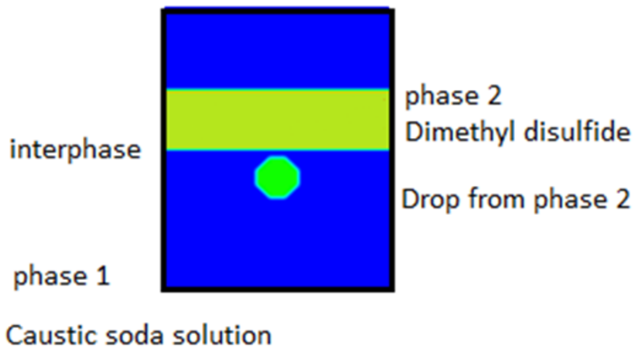

Figure 2. Computational domain, primary and boundary conditions used in simulations.

2.3. Estimation of coalescence times.

For modeling this phenomenon we considered the droplet position from the moment of movement and until it reaches the interface of two phases and it is investigated. The position is defined according to the initial position of the common interface of the two liquid phases (commonly referred to as the common interface). Proposed Time is defined according to the moment of reaching the initial position of the interfacing. In the simulation, the solution is saved after every 100 time steps.

The data saved is analyzed to check for the occurrence of the events of approach to the interface and drop interface coalescence. Simulations are repeated starting with the solution corresponding to the smaller time. The data are now saved after every 10 time steps and analyzed the event of approaching drainage or coalescence. The time step size is 0.00 25 s. The grid size increases the precision of modeling. The event of film drainage is illustrated in Figure 2 which shows the contours of the phase index at the time of the drainage event (t = 0.1000 s).

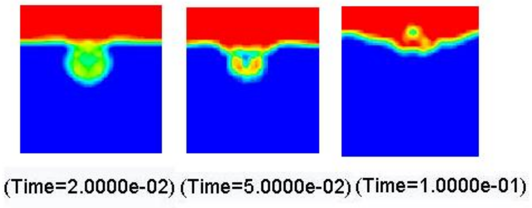

Figure 3. CFD simulation of drop coalescence

Selection of Grid Size and Validation. The simplicity of the computational domain allows us to use hexahedral grids. A constant time step size (0.001 s) is used in the simulations. Experimental data are derived based on factors that effecting coalescence time and design of the experiment. The experimental data for a drop of dispersed phase (dimethyl disulfide) with three diameters of 2, 5 and 7.5 mm passing through the interface between dimethyl disulfide and sodium hydroxide and coalescence with mother phase. The physical properties are shown in Table 1.

Table 1. Physical properties of all the liquids used in this work.

Composition	Density (g/L)			Viscosity (mPa.s)			Surface tension (mN/m)		
	20 ˚C	40 ˚C	60 ˚C	20 ˚C	40 ˚C	60 ˚C	20 ˚C	40 ˚C	60 ˚C
10% sodium hydroxide solution	1.109	1.099	1.089	1.88	1.25	0.82	78.62	74.57	70.52
12% sodium hydroxide solution	1.131	1.121	1.111	2.06	1.32	0. 86	79.87	76.05	71.69
14% sodium hydroxide solution	1.153	1.143	1.132	2.46	1.41	0. 91	81.27	77.65	72.97
Dimethyl disulfide	1.062	1.041	1.018	0.62	0. 49	0. 37	34.87	30.68	28.71

Several simulations are carried out to observe the effect of grid size on the coalescence time predicted by the CFD model for matching experimental results. We have grid the domain with the intervals of 0.1. Based on experimental and laboratory data, we record the density, viscosity and surface tension of each of the phases mentioned in the model. We measured the density by the DA-100M Density meter device manufactured by METLER TOLEDO company in Germany which works based on the electro-magnetically induced oscillationsof glass U-tube., and viscosity was measured via Viscometer Schot& CT72 device made in Germany and interfacial tension was measured by IFT 700 with pendant drop method. For quantitative validation, the predicted changes from the droplet position to time are compared with experimental data. This comparison is shown in Figure 3. A good match is observed between the results of CFD prediction and empirical data.

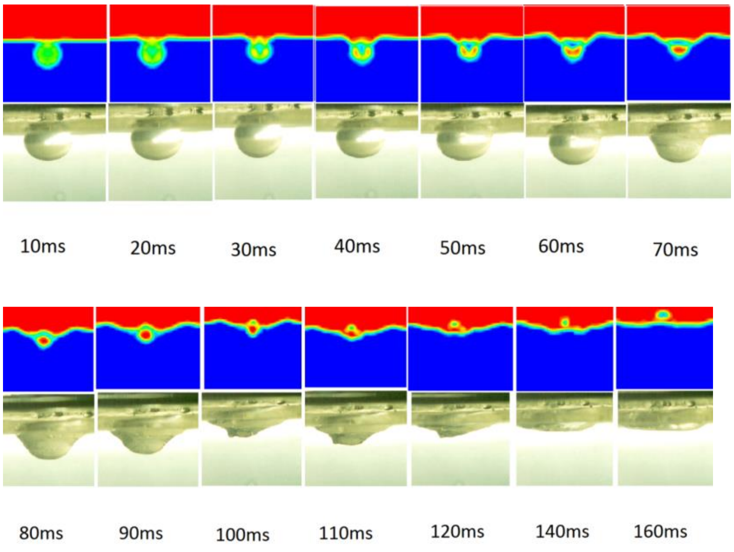

Figure 4. Comparison of the images obtained from experiments with the snapshots obtained from CFD simulation.

Figure 4 compares the images that were taken from the experiments with photographs taken from the CFD simulation. Phenomena such as the arrival of droplet into the interface, rupturing and drainage discharging, and eventually coalescence with mother phase at interface in both experimental result and the simulation of CFD. Phenomena such as approach to the interface even before the drop reaches the interface, elongation of the drop along the axis as it passes through the interface, The drop shape almost spherical in the heavier liquid to oblate ellipsoid in the lighter liquid, pulling up of the heavier liquid by the drop, and breakage of the thread are observed both in experiments as well as in CFD simulation. Quantitative and qualitative agreement between the predictions of the CFD model and experimental observations thus validates the CFD model. The validated CFD model is used for parametric analysis [14]. It must be mentioned that the drainage of the film is a complex phenomenon with very small-scale interactions becoming important when the film becomes too thin [6]. Therefore, to resolve film drainage, prohibitively fine grid is required which is not possible due to exorbitant computational demand. The grids used in this study, though not very fine, offer a good trade-off between the computational time and accuracy. Though there are many studies on CFD simulations of a drop or drop rising in a liquid [15-17].

3. RESULTS AND DISCUSSION

3.1 Design plan and parametric analysis.

The numbers of effective factors in this process are as follows: Diameter of the droplet, the concentration of caustic soda phase as the dispersed phase, Operating temperature, the length between release point and the contact surface, viscosity, density and interfacial tension. But the independent variable is the fourth one [18-20].

Each of these factors was considered at three levels in the design of the central composite in order to minimize the coalescence time of the Central composite design (CCD) dispersed-phase and mother phase on the contact surface. As mentioned above the goal of the research is to optimize the coalescence time. The Initial experiments were performed to determine the concentration of caustic soda, soda density, viscosity and surface tension. In designing the test for CCD design, 4 factors and three levels were considered to minimize the coalescence time of the CCD dispersed-phase and mother phase on the contact surface [21]. Thirty tests were run to study the desired results and six of them were run in the central point to estimate the error rate in empirical analysis. In order to reduce the possibility of error, each experiment was performed at least three times. The time it took for the drop to reach the contact surface and also the coalescence time with mother phase in the interface was captured by the high-speed camera. Besides this matter, The behavior of drops coalesscence with mother phase at the interface depends on various forces. The forces promoting the passage of the drop through the interface is the driving forces. The forces trying to prevent the passage of the drop through the interface is the restraining forces. The interfacial tension force is the important restraining force for increasing the coalescence time. The magnitude of the interfacial tension force depends on the interfacial tension between the two liquids. Thus, interfacial tension between the two liquids is an important parameter which affects the coalescence of drop with mother phase at interface. The main driving force is the inertial force corresponding to the velocity with which the drop approaches the interface. The density and viscosity are the parameters that effects on approach velocity and coalescence time. At the interface the drop senses a change of the surrounding liquid. Both density and viscosity of the liquid affect the rise velocity of a drop in the liquid. For a drop rising in a viscous liquid, the rise velocity is inversely proportional to the viscosity of the liquid and directly proportional to the density difference or the density of the liquid. In the parametric analysis we study the effect of the above mentioned important parameters on the phenomenon of the passage of the drop through the liquid−liquid interface .for better understanding the effect of each factor, we assumed that only one parameter is varied while other parameters are kept constant at their base values.

Specifically, we seek to understand how these parameters affect the coalescence time. This understanding is gathered by carrying out simulations in which only one of the parameters is varied while other parameters are kept constant at their base values. Such a freedom to vary only one of the parameters while keeping other parameters unchanged is compared with experiments and is generally accepted as one of the main advantages computations have over experiments [22]. Table 1 lists the physical properties base values of the parameters used in CFD simulations. There for we considered the following parameter that affecting the coalescence time in experiments Drop diameter , viscosity, Density, Interfacial tension concentration of continuous phase and we investigate the effect of each factors on coalescence time .

3.2. Effect of Drop Diameter on coalescence time.

Experiment and Simulations were carried out for different values of drop diameters ranging from 0.1 to 1 mm. The results obtained from the experiments and simulations are shown in Figure 5.

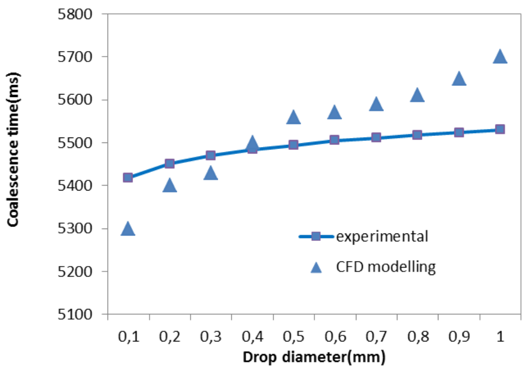

Figure 5. Comparison the effect of Drop diameter on coalescence time experimental and CFD modeling.

This is because the rise velocity (terminal settling velocity) of a drop reduces with a reduction in drop diameter. For the same reason, the slope of the drop position versus time curve is maximum for the largest drop. Shape of a rising drop is depending on a relative magnitude of gravitational, surface tension, inertial, and viscous forces. Shapes such as spherical, oblate spheroid, oblate ellipsoidal, etc. are possible depending on the values of Reynolds number and Eotvos number which characterizes the different forces acting on the drop [23]. Spherical shape is observed for small values of Reynolds and Eotvos numbers. In our simulations, the Reynolds number and Eotvos number is small in the heavier liquid. Hence the shape of the drop is almost spherical in the heavier liquid. Therefore, we considered a spherical drop raising in continuous phase. For the conditions of the simulations, the viscosity of the lighter liquid is 0.000549 kg/m s which is 2.5 times less than the viscosity of the heavier liquid. The density of the lighter liquid is about 0.94 times the density of the heavier liquid. Thus, the rise velocity of the drop which is inversely proportional to the continuous viscosity and directly proportional to the density of the external phase is significantly more in the lighter liquid than in the heavier liquid. At the interface, with reduction drop that is inside the heavy phase overall time coalescence will be reduced. Fig 5 shown that comparison of experimental results with CFD modeling for dimethyl disulfide drop separated from nozzle at sodium hydroxide (heavy phase) and raised up to the interface and finally coalescence with mother phase.

3.3. Effect of Interfacial tension on drop coalescence time.

As discussed earlier, the interfacial tension force is the main restraining force that tries to resist the passage of the drop through the interface.

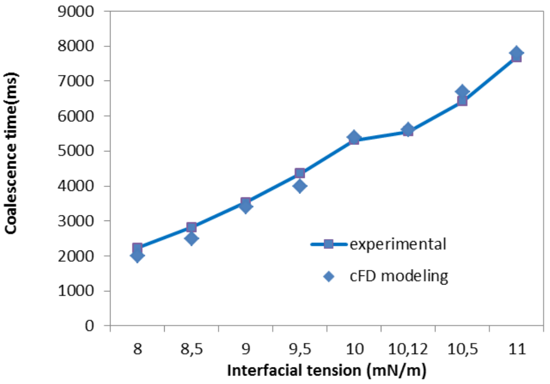

Figure 6. The effect of interfacial tension on coalescence time.

The higher the interfacial tension, the more is the resistance. Simulations were carried out for different values of interfacial tension between the two liquids while keeping the other physical properties and drop diameter at the base values. The interfacial tension was varied in the range from 8 to 11 mN/m. This is because the drop spends different time at the interface for different values of interfacial tension. Fig 6 suggests that the lower the interfacial tension, the faster is the movement of the drop through the liquid–liquid interface.

Retention time is found to reduce monotonically with increase in interfacial tension. This is because a higher interfacial tension causes faster retraction of the heavier liquid pulled up by the drop. That the retention time reduces with increase in interfacial tension the retardation of the drop at the interface increases with increase in interfacial tension. For high interfacial tension, the drop almost stops rising. It sits at the interface waiting for the film to drain and coalescence with mother phase. This causes the overall process of the coalescence of the drop to slow down with an increase in interfacial tension despite reduction in retention time with increase in interfacial tension. As observed in the case of dependence of the drainage time on drop diameter, the drainage time does not change monotonically with interfacial tension. With the increase in interfacial tension, the film drainage should become faster. But due to flattening of the drop with increase in interfacial tension, the volume of the heavier liquid trapped in the film also increases, which can take more time to drain. Because of these two counteracting factors, the drainage time does not change monotonically with change in interfacial tension.

3.4. Effect of viscosity Ratio on drop coalescence time.

Experiment work and Simulations were carried out to study the effect of the viscosity ratio of the two liquids on the phenomenon of drop coalescence with mother phase at interface. Fig.7 summarizes the results obtained from these simulations and experiment.

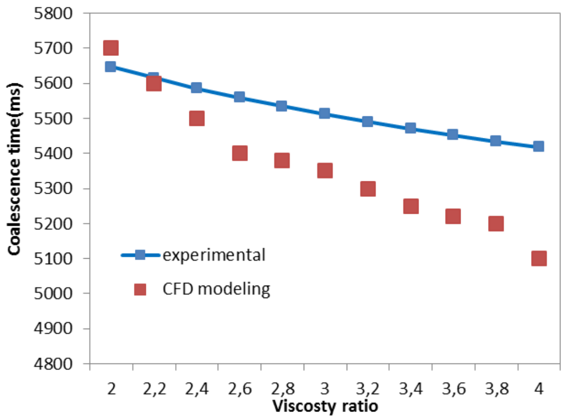

Figure 7. The effect of viscosity Ratio on coalescence time.

Here the viscosity ratio means the ratio of viscosities of the heavier liquid and the lighter liquid. Unless otherwise stated, viscosity ratio is varied by varying the viscosity of the heavier liquid keeping the viscosity of the lighter liquid constant. As can be seen, when the viscosity ratio decreases, i.e., the viscosity of the heavier liquid reduces, the movement of the drop becomes slower in the heavier liquid. With a decrease in the viscosity ratio, the drop reaches the interface with a higher velocity and coalescence with mother phase at the interface faster. The parts of the curves corresponding to the movement of the drop in the lighter liquid are parallel to each other. This is because the lighter liquid is essentially the same in these simulations and experiments, and hence the rise velocity of the drop in the lighter phase is predicted to be the same. As the viscosity ratio increased to large values, the drainage time and retention time increase steeply. A higher viscosity ratio corresponds to a higher viscosity of the heavier liquid. Thus, at lower viscosity ratio, the velocity with which the drop approaches the interface reduces. Therefore, for a lower viscosity ratio the inertial force responsible for the drainage of the film is less. With a reduction in the viscosity ratio, the viscosity of the film also increases making the drainage of the film slower. Both these factors reduced approach velocity of the drop and increased the viscosity of the film combine to steeply increase the drainage time when the viscosity ratio is reduced below unity. As the viscosity ratio is decreased, the heavier phase becomes less viscous. This should enhance the approach velocity of the drop, and hence the drainage of the film should be faster with a decrease in the viscosity ratio.

This should cause drainage time to continuously reduce with increase in viscosity ratio. The simulation results, however, show that the drainage time eventually becomes constant with continued increase in the viscosity ratio. With an increase in the viscosity ratio, the shape of the drop when it reaches the interface becomes flatter. This traps more of the heavier phase in the film causing the drainage time to increase. This probably balances the effect of the enhanced approach velocity causing the drainage time to eventually become constant with continued increase in the viscosity ratio. The steep increase in retention time for a viscosity ratio less than unity can be attributed to a steep reduction in the velocity with which the drop approaches the interface.

3.5. Effect of Density Difference on drop coalescence time.

The density difference between the two liquids may also affect the coalescence time of the drop.

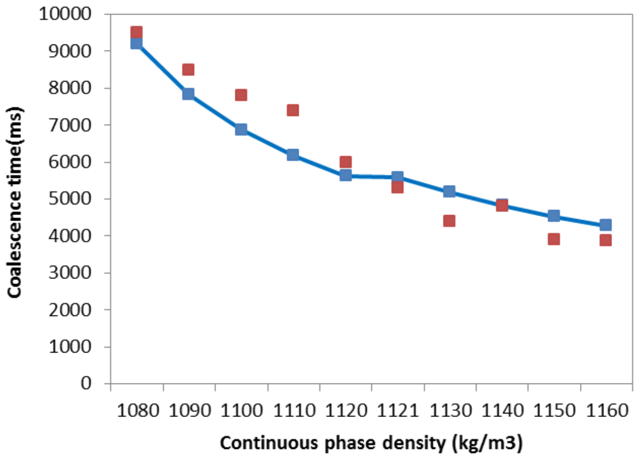

Figure 8. The effect of continuous phase density on the coalescence time.

To understand the role played by the density difference, experiment and simulations were carried out for different values of the density difference between the two liquids. Unless otherwise stated, density difference was varied by varying the density of the heavier liquid, keeping the density of the lighter liquid constant. Fig 8 summarizes the results obtained from these simulation and experimental results.

As the density difference increases, the density of the heavier liquid increases. This causes an increase in the rise velocity of the drop in the heavier liquid. Since for higher density difference the approach velocity of the drop is higher, it passes through the interface faster. The retention time is found to reduce monotonically with increase in density difference. Since on increasing the density difference the density of the heavier phase increases, the gravitational force acting on the heavier liquid pulled up by the drop increases with increase in density difference. This causes faster retraction of the heavier liquid pulled up by the drop causing the retention time to reduce with increase in density difference. The drainage time is also found to reduce monotonically with an increase in density difference. This can be attributed to enhanced driving force for the film drainage due to higher approach velocity of the drop when density difference is increased by increasing the density of the heavier liquid.

4. CONCLUSION

The phenomenon of drop coalescence with mother phase at the interface between two immiscible liquids has been studied by using CFD simulations and experimentally. The liquids considered are Newtonian liquids. Flow regime is laminar. In the simulation the VOF method has been used for tracking the interfaces between different fluids and for experiments has been used from design experiment and high speed camera. Validation has been carried out using a comparison between two methods. The validated computational approach has been used to study the effects of drop diameter and important physical properties such as interfacial tension, viscosity Ratio, and density difference on the phenomenon of drop coalescence. The following highlights are the important conclusions were derived from both results:

- With increase in drop diameter, the overall drop coalescence time with morther phase at liquid−liquid interface increased and coalescence becomes slower. Beside this matter drainage time of the drop increases with increase in drop diameter.
- The overall drop coalescence time with mother phase will be increased with increase in interfacial tension. Despite this matter retention time reduce with increase in interfacial tension.
- With reducing (ratio of the viscosity of the heavier liquid and the lighter liquid) the drop coalescence time with mother phase increase. And for viscosity less than the retention time almost independent of viscosity ratio.
- The drop coalescence time by increasing density of heavier liquid will be decreased. With increasing in density difference (at a constant density of light liquid) the drop coalescence with mother phase at interface becomes faster. The retention time and the drainage time is reduced with the increase in density difference between the two liquid phases.

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