



## Original Article

## Drop formation at submerged nozzles: Comparison of aqueous dispersed and organic dispersed cases for TBP-dodecane and nitric acid system

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## ABSTRACT

Understanding the phenomena of formation of single drops is necessary to understand the hydrodynamics in solvent extraction equipment which are used for separation of nuclear materials. In this work, the phenomena of aqueous phase and organic phase drop formation at submerged nozzles are compared by conducting experiments with 30%TBP (v/v) in dodecane as the organic phase and nitric acid as the aqueous phase. Two different nozzles and three different nitric acid concentrations are used. For each nozzle and nitric acid concentration, velocity of the dispersed phase is varied. Drops of aqueous phase formed at downward oriented nozzles submerged in organic phase are observed to be smaller than the drops of organic phase formed at upward oriented nozzles submerged in aqueous phase. Correlations to estimate drop diameter are proposed.

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## 1. Introduction

Two-phase flows are common in nuclear industry [1,2]. Solvent extraction, which also involves liquid-liquid two-phase flow, is extensively used for separation of nuclear materials [3,4]. Different designs of solvent extraction equipment are used in the nuclear industry [5–7]. All these designs generate swarm of small drops, size of which affects specific interfacial area for interphase mass transfer. Effective settling of the liquids after mass transfer also depends on the drop size. Therefore, to estimate mass transfer and settling rates, estimation of drop size is required. There has been significant progress in estimating drop size by population balance modelling [8–12]. However, studies on single drops are still required to perfect the breakage and coalescence kernels used in the population balance models. Though, in a solvent extraction equipment the drop size is mainly governed by the degree of

turbulence, inlet drop size distribution may also affect the drop size and dispersed phase holdup inside the equipment [13]. Thus, understanding phenomena of drop formation on distributor plates and submerged nozzles is essential.

Drop formation at upward oriented submerged nozzles has been studied extensively [14–16]. However, drop formation at downward oriented nozzles, which is much less studied, is equally important. A solvent extraction equipment can operate in either organic dispersed or aqueous dispersed mode. For example, in a pulsed perforated plate column, generally, the aqueous phase which preferentially wets the plate material, usually stainless steel, is the continuous phase. However, in some cases the aqueous phase needs to be dispersed [17]. In such cases, nozzle plates having holes with small nozzle like protrusion oriented downward, are used. When the aqueous phase passes through these nozzle-like downward oriented protrusions, it does not spread along the plate and gets dispersed. Thus, drop formation at downward oriented nozzles when aqueous phase is the dispersed phase is as important as drop formation at upward oriented nozzles when the organic phase is the dispersed phase. Considering this, aqueous phase drop formation at downward oriented nozzles and organic phase drop formation at upward oriented nozzles have been compared in this work. The liquid – liquid system used is 30% (v/v) tri-butyl

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phosphate (TBP) in dodecane as the organic phase and nitric acid of varying concentrations (0.01 N, 1 N, 3 N) as the aqueous phase. The system is chosen due to its relevance in the nuclear industry [3,18]. Table 1 summarizes some of the previous studies on drop formation at submerged nozzles. It shows that the studies with liquid-liquid systems relevant to nuclear fuel cycle are very few. The studies on comparison of organic phase drop formation at upward oriented nozzles and aqueous phase drop formation at downward oriented nozzles are rare. In the present study, we intend to address these gap areas. In this work, experiments are conducted to determine drop diameter, drop detachment height and drop detachment time for varying dispersed phase velocity, nozzle diameter and nitric acid concentration. High speed imaging is used to capture drop formation.

## 2. Experimental setup and procedure

Fig. 1 shows the experimental setup for aqueous phase drop formation at a downward oriented nozzle. The setup for organic phase drop formation was described in our previous study [27]. Stainless steel nozzles ( $d_N = 1.91$  mm, 3.04 mm) are used.  $d_N$  represents the inner diameter of the nozzle. In our previous study on organic phase drop formation at upward oriented nozzles, three different nozzles ( $d_N = 1.91$  mm, 3.04 mm, 4.88 mm) were used. However, during the experiments on aqueous phase drop formation at 4.88 mm inner diameter nozzle, a continuous stream of aqueous phase was obtained. Thus, in this study, we report the results of the experiments conducted with 1.91 mm and 3.04 mm inner diameter nozzles only. Aqueous phase is nitric acid of different concentrations (0.01 N, 1 N and 3 N). 30% (v/v) TBP in dodecane is the organic phase. The phases are mutually saturated before conducting the experiments. Physical properties of the liquid-liquid systems are given in Table 2.  $\rho$  is the density,  $\mu$  is the viscosity,  $\Delta\rho$  is the density difference between the aqueous phase and organic phase.  $\gamma$  is the interfacial tension. The glass column is filled with organic phase. Nitric acid is pumped by a syringe pump through the nozzle at a desired flow rate. Drop formation is captured using high speed camera. Average volume of drops and diameter are obtained by using the time of formation of 10 drops and flow rate of the dispersed phase. Nozzle having inner diameter equal to 1.91 mm is referred to as Nozzle-1. Nozzle having inner

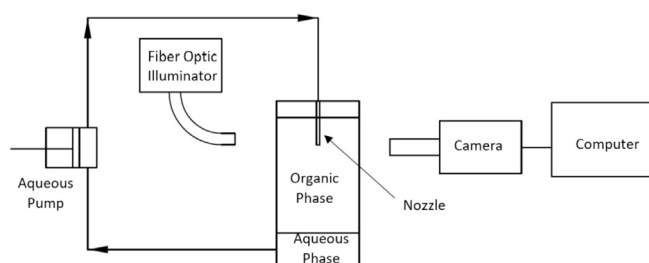


Fig. 1. Schematic diagram of the experimental setup for aqueous phase drop formation at downward oriented nozzle.

diameter equal to 3.04 mm is referred to as Nozzle-2.

## 3. Results and discussion

### 3.1. Visualization of drop formation phenomenon

Figs. 2 and 3 show typical images of organic phase drop formation at upward oriented nozzle and aqueous phase drop formation at downward oriented nozzle for the Two-phase System – I and 3.04 mm inner diameter nozzle. For organic phase drop formation, up to 45.9 mm/s velocity, the drop is attached to its bulk phase through a neck. The dispersed phase flows into the drop through the neck. The shape of the neck is concave. This regime of drop formation is called as dripping or necking regime. The thinning of the neck finally leads to detachment of the drop. As the velocity increases, jetting sets in and the shape of the neck becomes convex. Jet length (drop detachment length) increases with increase in velocity. At the maximum velocity used in the experiments, the onset of jet instability is observed. At small velocities, drop size is governed by the balance between buoyancy and interfacial tension forces. At higher velocities, the momentum and drag forces also contribute to the force balance.

As observed in Fig. 3, aqueous phase drops getting formed at downward oriented nozzle are almost spherical and attached through a neck to the nozzle for the entire range of the velocity covered in the experiments. At higher velocities, jetting is observed for downward oriented nozzle also. However, the jet is like a falling

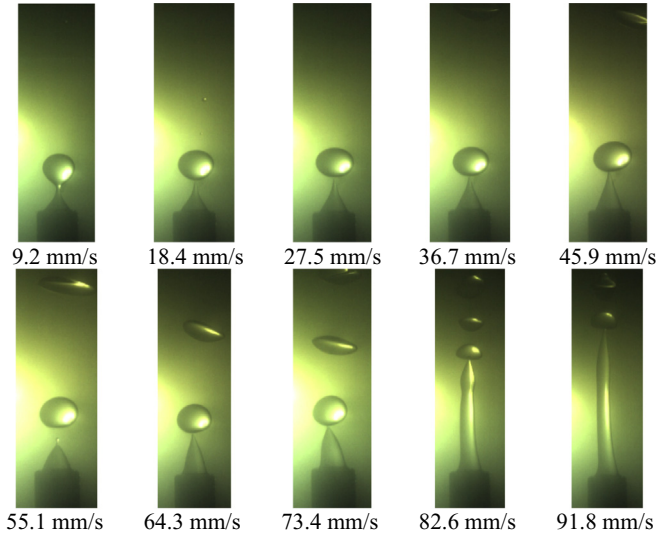
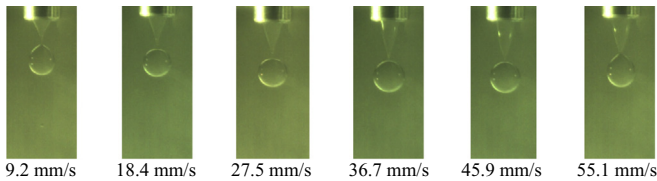
Table 1  
Summary of some previous studies on drop formation on nozzles/capillaries submerged in quiescent continuous phase.

Reference	Dispersed phase	Continuous phase	Nozzle/Capillary orientation
[14]	heptane, butyl alcohol, (55% carbon tetrachloride and 45% heptane), paraffin oil, (90% paraffin oil and 10% heptane), (80% paraffin oil and 20% heptane), (70% paraffin oil and 30% heptane), benzene, (95% benzene and 5% acetone)	water, glycerine, (90% glycerine and 10% water), (80% glycerine and 20% water), (70% glycerine and 30% water), (50% glycerine and 50% water), (95% water and 5% acetone)	Upward
[16]	heptane, butyl alcohol, (55% carbon tetrachloride and 45% heptane), paraffin oil, (90% paraffin oil and 10% heptane), (80% paraffin oil and 20% heptane), (70% paraffin oil and 30% heptane), benzene, (95% benzene and 5% acetone)	water, glycerine, (90% glycerine and 10% water), (80% glycerine and 20% water), (70% glycerine and 30% water), (50% glycerine and 50% water), (95% water and 5% acetone)	Upward
[19]	benzene, (benzene + alkaterge), (motor oil + kerosene), water, carbon tetrachloride	water, (water + sucrose), carbon tetrachloride, benzene, water	Upward & downward
[20]	cottonseed oil, water	water, benzene, carbon tetrachloride	Upward
[21]	benzene	(water + lissapol) and glycerine	Upward
[22]	water	(40% toluene and 60% paraffin oil), amylalcohol	Downward
[23]	kerosene	distilled water	Upward
[24]	carbon tetrachloride, bromobenzene, chlorobenzene, nitrobenzene	carboxymethylcellulose in water (0.50 wt%, 1.00 wt%, 1.50 wt%, 2.00 wt%), polyacrylamide in water (0.05 wt%, 0.10 wt%, 0.15 wt%, 0.20 wt%), polyvinylpyrrolidone in water (2.00 wt%, 3.00 wt%, 4.00 wt%)	Upward
[25]	0.16% xanthan gum in 80:20 glycerol/water by volume	9.8 P polydimethylsiloxane (PDMS) oil.	Downward
[26]	toluene	water	Upward
[27]	30% TBP in dodecane	nitric acid of different concentrations	Upward

**Table 2**

Physical properties (at 25 °C) of the two-phase systems used in the experiments.

Test System		$\rho$ (kg/m <sup>3</sup> )	$\mu$ (cP)	$\Delta\rho$ (kg/m <sup>3</sup> )	$\gamma$ (mN/m)
Two-phase System – I					
Aqueous	0.01 N nitric acid	1017	0.81	194	9.38
Organic	30% (v/v) TBP in dodecane	823	1.54		
Two-phase System – II					
Aqueous	1 N nitric acid	1032	0.83	208	9.62
Organic	30% (v/v) TBP in dodecane	824	1.61		
Two-phase System – III					
Aqueous	3 N nitric acid	1094	0.91	262	10.99
Organic	30% (v/v) TBP in dodecane	832	1.70		

**Fig. 2.** Snapshots of organic phase drop formation at upward oriented 3.04 mm inner diameter nozzle (Nozzle-2) for different velocities of the organic phase at the nozzle. Continuous phase is 0.01 N nitric acid.**Fig. 3.** Snapshots of aqueous phase drop formation at downward oriented nozzle of 3.04 mm diameter (Nozzle-2) for different velocities of the aqueous phase (0.01 N nitric acid) at the nozzle.

stream of the dispersed phase, length of which goes beyond the field of view of the camera. This stream directly impinges the interface between the aqueous and organic phases at the bottom of the glass vessel. This jetting is different from jetting observed in case of organic phase drop formation at upward oriented nozzles in which jet length is finite.

### 3.2. Comparison of aqueous phase and organic phase drop formation phenomena

#### 3.2.1. Comparison of drop diameter

Fig. 4 shows that for the same dispersed phase velocity through the nozzle ( $U_N$ ), the diameter ( $d$ ) of the aqueous phase drops formed at downward oriented nozzles is less than the diameter of the aqueous phase drops formed at upward oriented nozzles. According to the two-stage drop formation model, the drop remains

attached to the nozzle through a neck till the restraining forces, the forces that try to keep the drop attached to its bulk phase, are in balance with the disrupting forces, the forces that are responsible for detachment of drop from its bulk phase [14–16,21,28,29]. The restraining forces are interfacial tension force ( $F_I$ ), drag force ( $F_{DR}$ ) and inertia force ( $F_{INER}$ ). The disrupting forces are buoyancy force ( $F_B$ ) and momentum or kinetic force ( $F_M$ ) [23]. During this period, dispersed phase continues to flow into the drop through the neck, causing drop volume to increase till the time of drop break off. The different forces are represented by Eqns (1)–(5).

$$F_B = \frac{4}{3} \pi r^3 |\rho_c - \rho_d| g \quad (1)$$

$$F_M = \frac{Q_N^2 \rho_d}{\pi r_N^2} \quad (2)$$

$$F_I = 2 \pi r_N \gamma \quad (3)$$

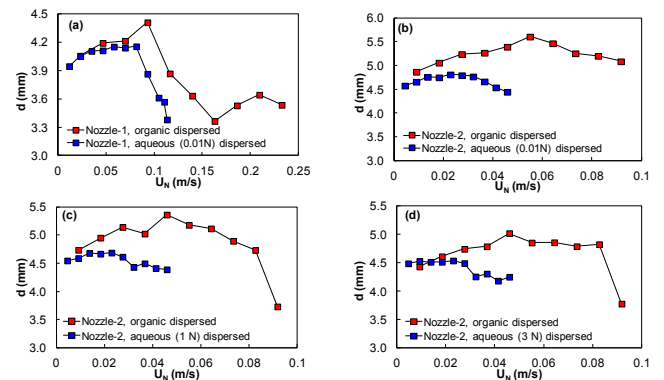
$$F_{DR} = \frac{1}{2} C_D A \rho_c U_d^2 \quad (4)$$

$$F_{INER} = \frac{m_v Q_N^2}{12 \pi r^2} \quad (5)$$

Drop starts leaving the nozzle when the disrupting forces become more compared to restraining forces as given by Eq (6).

$$F_B + F_M > F_I + F_{DR} + F_{INER} \quad (6)$$

$r$  and  $U_d$  are the drop radius and drop expansion velocity just at the

**Fig. 4.** Variation of drop diameter with dispersed phase velocity for organic phase drop formation at upward oriented nozzle and aqueous phase drop formation at downward oriented nozzle. (a) Nozzle-1, 0.01 N nitric acid (b) Nozzle-2, 0.01 N nitric acid (c) Nozzle-2, 1 N nitric acid (d) Nozzle-2, 3 N nitric acid. The experimental data for the organic dispersed case are from our previous study [27].

time of drop getting detached from the nozzle with the formation of a neck.  $\rho_c$  and  $\rho_d$  are the densities of the continuous phase and dispersed phase, respectively.  $r_N$  is the nozzle radius.  $Q_N$  is the dispersed phase flow rate.  $C_D$  is drag coefficient.  $\gamma$  is the interfacial tension.  $A$  is the projected area.  $g$  is acceleration due to gravity. The virtual mass  $m_v$  used in Eq. (5) is defined by Eqs. (7) and (8).

$$m_v = \rho_d + \alpha \rho_c \quad (7)$$

$$\alpha = \left(\frac{1}{2}\right) \left[1 + \frac{3r^3}{8s^3}\right] \quad (8)$$

$S$  is the distance from the centre of the drop to the wall of the column filled with the continuous phase. In the present study  $r \ll S$ , hence  $\alpha$  can be taken as 0.5. To compare the forces during organic phase and aqueous phase drop formation, let us consider the force balance on a drop of radius  $r$ , getting formed at a nozzle of radius  $r_N$ , at the dispersed phase volumetric flow rate  $Q_N$  and drop expansion velocity  $U_d$ . For the same diameter of the drop,  $F_B$  and  $F_I$  are the same irrespective of the organic phase being dispersed or aqueous phase being dispersed. The net disrupting force for downward drop formation ( $F_{NDFD}$ ) in which aqueous phase is dispersed can be expressed by Eq. (9)

$$F_{NDFD} = F_B + F_M - F_I - F_{DR} - F_{INER} \quad (9)$$

Using Eqns. (1)–(8), Eq. (9) for the case of aqueous phase drop formation at downward oriented nozzle can be written as

$$F_{NDFD} = \frac{4}{3} \pi r^3 |\rho_o - \rho_a| g + \frac{Q_N^2 \rho_a}{\pi r_N^2} - 2 \pi r_N \gamma - \frac{1}{2} C_D A \rho_o U_d^2 - \frac{(\rho_a + 0.5 \rho_o) Q_N^2}{12 \pi r^2} \quad (10)$$

Similarly, the net disrupting force during organic phase drop formation at upward oriented nozzle ( $F_{NDFU}$ ) can be written as

$$F_{NDFU} = \frac{4}{3} \pi r^3 |\rho_a - \rho_o| g + \frac{Q_N^2 \rho_o}{\pi r_N^2} - 2 \pi r_N \gamma - \frac{1}{2} C_D A \rho_a U_d^2 - \frac{(\rho_o + 0.5 \rho_a) Q_N^2}{12 \pi r^2} \quad (11)$$

Thus, the difference between net disrupting force for downward aqueous phase drop formation and upward organic phase drop formation can be written as

$$F_{NDFD} - F_{NDFU} = \frac{Q_N^2 (\rho_a - \rho_o)}{\pi} \left( \frac{1}{r_N^2} - \frac{0.5}{12 r^2} \right) + \frac{1}{2} C_D A (\rho_a - \rho_o) U_d^2 \quad (12)$$

Since,  $\rho_a > \rho_o$  and  $r_N < r$ , the net disrupting force on a downward forming aqueous phase drop is more than the net disrupting force on an upward forming organic phase drop of identical size. Thus, for the same phase system, for identical flow rate of the dispersed phase and nozzle diameter, an aqueous phase drop forming at a downward oriented nozzle should detach faster and its diameter should be smaller compared to an organic phase drop forming at an upward oriented nozzle. This explains the experimental results shown in Fig. 4. Fig. 4 also shows that for aqueous dispersed as well as organic dispersed case the drop diameter does not vary monotonically with dispersed phase velocity.

### 3.2.2. Comparison of drop detachment height

Fig. 5 shows that the drop detachment height ( $L$ ) is more for aqueous phase drop formation for all nozzle diameters and nitric

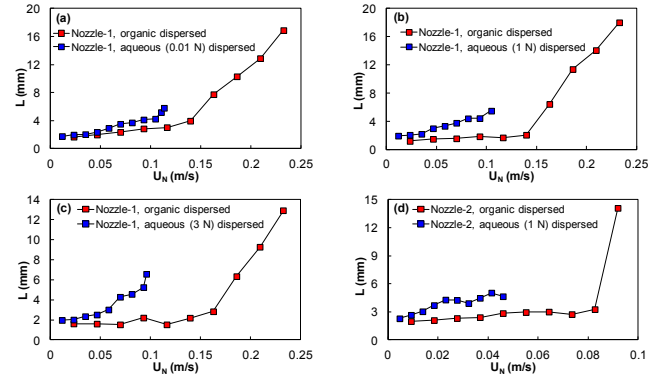


Fig. 5. Variation of drop detachment height with dispersed phase velocity for organic phase drop formation at upward oriented nozzle and aqueous phase drop formation at downward oriented nozzle (a) Nozzle-1, 0.01 N nitric acid (b) Nozzle-1, 1 N nitric acid (c) Nozzle-1, 3 N nitric acid (d) Nozzle-2, 1 N nitric acid. The experimental data for the organic dispersed case are from our previous study [27].

acid concentrations. For organic phase drop formation, the drop detachment height first increases moderately and then steeply with continued increase in dispersed phase velocity. This change in the slope signifies distinct transition from necking to jetting regime. Fig. 5 also shows that for lower dispersed phase velocities, the rate of increase of drop detachment height with increase in dispersed phase velocity is more for aqueous phase drop formation than for organic phase drop formation. However, for aqueous phase drop formation the transition from necking to jetting is not clearly observed. It may be noted that, in Fig. 5, the range of dispersed phase velocity for aqueous phase drop formation is much less compared to organic phase drop formation. This is because of earlier transition from necking to jetting in case of aqueous phase drop formation. However, the jet for aqueous dispersed case was like a free falling liquid stream and it extended beyond the field of view of the camera. Due to this, the length of the jet for aqueous dispersed case could not be determined. Thus the points of large drop detachment height in jetting regime for aqueous dispersed case are not there in Fig. 5. The figure nevertheless shows that for the same velocity drop detachment height is more for aqueous dispersed case compared to organic dispersed case.

### 3.2.3. Comparison of drop detachment time

Fig. 6 shows that for all nozzle diameters and nitric acid concentrations, the drop detachment time ( $T$ ) is less for aqueous phase drop formation than the same for organic phase drop formation. Since the drop diameter is less for the aqueous phase drop formation, as observed in Fig. 4, the drops have to break off at a higher frequency and drop detachment time has to be less for the aqueous dispersed case for a given flow rate of the dispersed phase and nozzle diameter. For both cases, the drop detachment time reduces monotonically with increase in dispersed phase velocity.

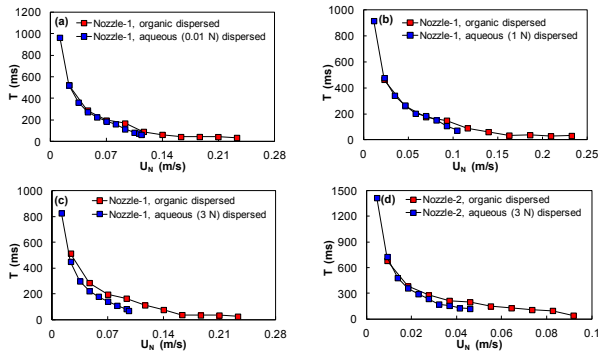
### 3.3. Drop formation at downward oriented nozzles

The phenomenon of organic phase drop formation at upward oriented nozzles was described in our previous study [27]. In this section, we provide the results of the experiments on aqueous phase drop formation at downward oriented nozzles.

#### 3.3.1. Effect of dispersed phase velocity and nozzle diameter on drop diameter

As nozzle diameter is increased, the restraining force due to interfacial tension increases, as can be seen from Eq. (3). Similarly,



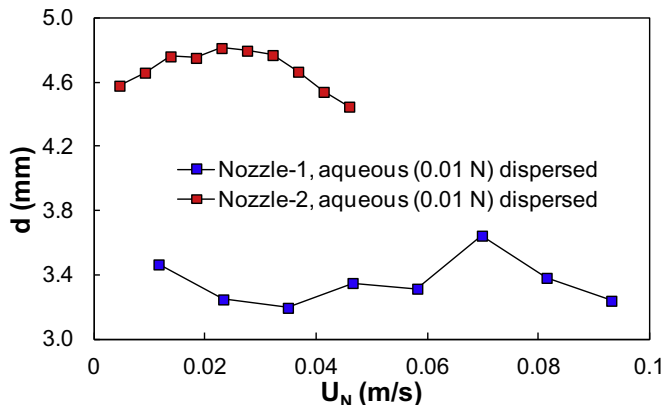


**Fig. 6.** Variation of drop detachment time with dispersed phase velocity for organic phase drop formation at upward oriented nozzle and aqueous phase drop formation at downward oriented nozzle (a) Nozzle-1, 0.01 N nitric acid (b) Nozzle-1, 1 N nitric acid (c) Nozzle-1, 3 N nitric acid (d) Nozzle-2, 3 N nitric acid. The experimental data for the organic dispersed case are from our previous study [27].

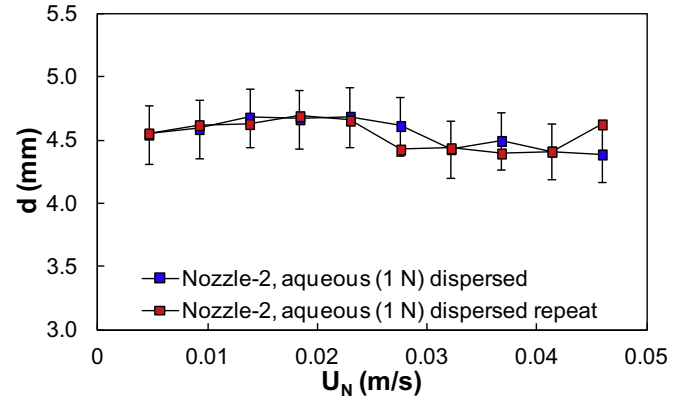
for the same dispersed phase velocity, the restraining force due to inertia scales as square of nozzle diameter. Thus for the same dispersed phase velocity, the restraining forces are expected to be larger for a nozzle having larger diameter. Larger restraining forces for a larger nozzle diameter would mean that the drop must grow to a larger volume for the disrupting forces to overcome the restraining forces. Thus for the same dispersed phase velocity, drop diameter is larger for a larger nozzle diameter, as observed from Fig. 7. The trends of variation of drop diameter with nozzle velocity and nozzle diameter for aqueous phase drop formation at downward oriented nozzles are the same as that of organic phase drop formation at upward oriented nozzles reported in our previous study [27]. Repeatability of experiments was also ascertained and found to be very good with the deviation between drop diameters obtained for two sets of experiments being less than 5%, as shown in Fig. 8.

### 3.3.2. Effect of dispersed phase velocity and nozzle diameter on drop detachment height

Fig. 9 shows that the drop detachment height increases with increase in nozzle velocity and nozzle diameter for aqueous phase drop formation at downward oriented nozzles. The same trend was observed for organic phase drop formation at upward oriented nozzles [27]. With an increase in dispersed phase velocity, the dispersed phase is able to penetrate farther into the quiescent continuous phase. Thus, the drop detachment height increases



**Fig. 7.** Variation of drop diameter with dispersed phase velocity and nozzle diameter for aqueous phase drop formation at downward oriented nozzles.



**Fig. 8.** Result of the experiments conducted to check the repeatability of the experimental data. The error bars are  $\pm 5\%$  error bars around the first set of data.

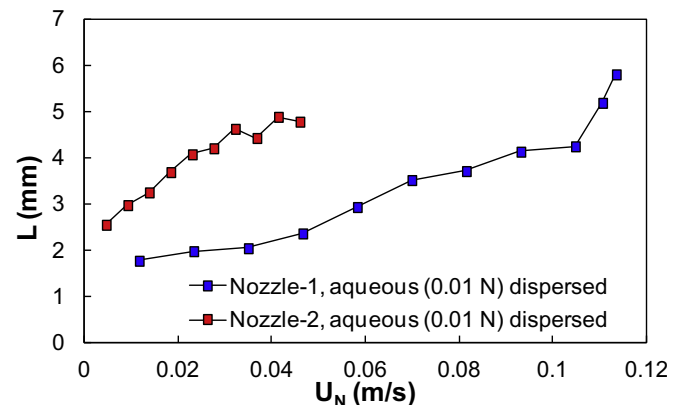
with an increase in dispersed phase velocity. As the nozzle diameter increases, the thickness of the column that connects the drop being formed to its bulk phase increases. A thicker column penetrates deeper into the quiescent continuous phase before it thins out and breaks. This causes drop detachment height to increase with increase in nozzle diameter.

### 3.3.3. Effect of dispersed phase velocity and nozzle diameter on drop detachment time

Fig. 10 shows the effect of dispersed phase velocity and nozzle diameter on drop detachment time for aqueous phase drop formation. Drop detachment time reduces with increase in dispersed phase velocity and with increase in nozzle diameter. The same trends were observed for the organic phase drop formation at upward oriented nozzles also [27].

### 3.3.4. Effect of nitric acid concentration

Fig. 11 shows that the diameter of the aqueous phase drops formed at downward oriented nozzles reduces with an increase in nitric acid concentration. The same trend was observed for organic phase drop formation at upward oriented nozzles [27]. The interfacial tension between the aqueous phase and organic phase for the three test systems are almost the same but the density of the aqueous phase (dispersed phase in case of the downward oriented nozzles) increases with increase in nitric acid concentrations. Thus, the resultant of buoyancy and gravitational force which acts downwards and hence helps drop detachment at downward



**Fig. 9.** Variation of drop detachment height with dispersed phase velocity and nozzle diameter for aqueous phase drop formation at downward oriented nozzles.

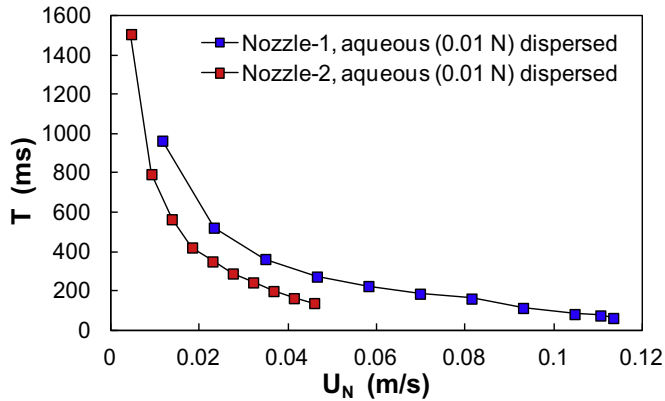


Fig. 10. Variation of drop detachment time with dispersed phase velocity and nozzle diameter for aqueous phase drop formation at downward oriented nozzles.

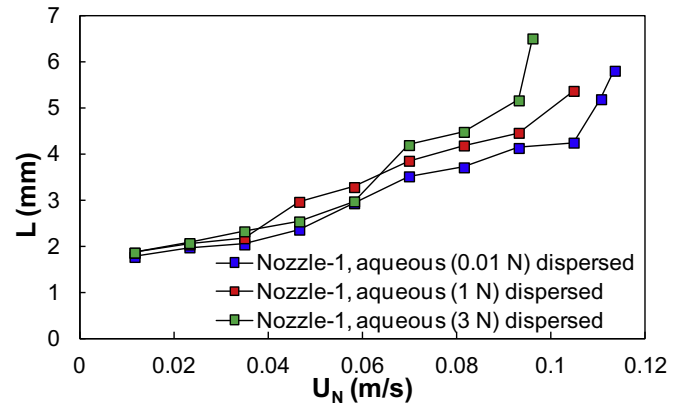


Fig. 12. Effect of nitric acid concentration and dispersed phase velocity on drop detachment height for aqueous phase drop formation at downward oriented nozzles.

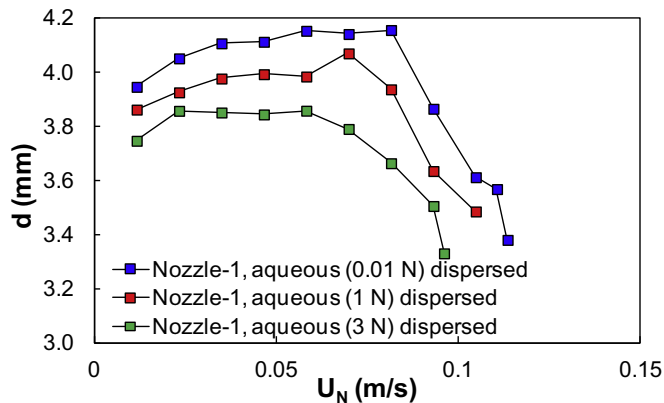


Fig. 11. Effect of nitric acid concentration and dispersed phase velocity on diameter of aqueous phase drops formed at downward oriented nozzles.

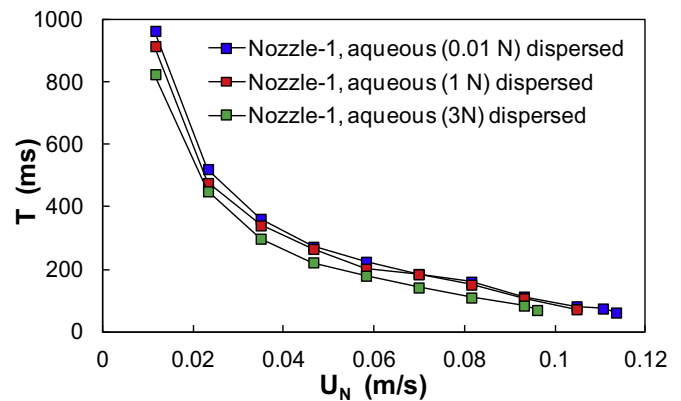


Fig. 13. Effect of dispersed phase velocity and nitric acid concentration on drop detachment time for aqueous phase drop formation at downward oriented nozzles.

oriented nozzle is more when nitric acid concentration is more. This causes drop diameter to reduce with an increase in nitric acid concentration in the aqueous phase for the same nozzle velocity and nozzle diameter.

Fig. 12 shows the effect of nitric acid concentration on drop detachment height for aqueous phase drop formation at downward oriented nozzles. The experimental trends suggest that for lower dispersed phase velocities there is no monotonic trend of variation of drop detachment height with nitric acid concentration. At higher dispersed phase velocities, drop detachment height increases with increase in nitric acid concentration. For organic phase drop formation at upward oriented nozzles also, there was no monotonic trend of variation of drop detachment height with nitric acid concentration [27].

Fig. 13 shows that the drop detachment time reduces slightly with increase in nitric acid concentration. For the same dispersed phase velocity and nozzle diameter, the flow rate of the dispersed phase is also the same. Since drop diameter reduces with increase in nitric acid concentration, for the same flow rate of the dispersed phase, the drop formation time must be smaller for a higher nitric acid concentration. The same trend of variation of drop detachment time with nitric acid concentration was observed for organic phase drop formation at upward oriented nozzles [27].

### 3.4. Correlations of drop diameter

The previously reported correlation to estimate drop diameter

for nitric acid - 30%TBP in dodecane system for organic phase drop formation at upward oriented nozzles is given by Eq. (13) [27].

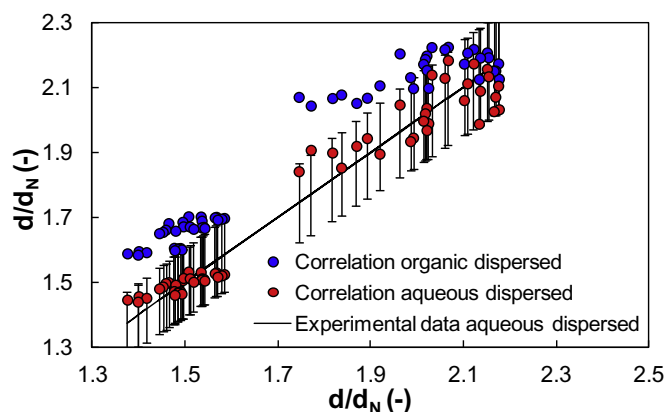
$$\frac{d}{d_N} = \frac{2.1962}{Eo} + 0.76739 \exp(-0.3976 Fr) - \frac{0.82523}{Eo^2} \quad (13)$$

Based on the experiments reported in this study, the correlation of drop diameter for aqueous phase drop formation at downward oriented nozzles as obtained by using Lab Fit Curve Fitting Software is given by Eq. (14) [30]. The functional form of the correlations for the two cases is the same.  $d_N$  is the nozzle diameter.

$$\frac{d}{d_N} = \frac{0.90594}{Eo} + 1.0665 \exp(-0.44353 Fr) - \frac{0.055946}{Eo^2} \quad (14)$$

The average absolute relative error in the fit of Eq. (14) is 2.55%. The maximum error is 7.81%. The parity plot is shown in Fig. 14. It is evident that the  $d/d_N$  values obtained by the correlation for organic phase drop formation are more than the  $d/d_N$  values obtained by the correlation for aqueous phase drop formation. Eötvös number ( $Eo = |\rho_c - \rho_d| g d_N^2 / \gamma$ ) and Froude number ( $Fr = U_N^2 / g d_N$ ) used in the correlations account for all the forces that affect the drop formation phenomenon. The ranges of Eötvös number and Froude number for the correlation of Eq. (14) are 0.74–1.97 and 0.0007 to 0.69, respectively.

Fig. 14 shows the  $d/d_N$  values predicted by the correlation for aqueous dispersed case comprise of two groups. In the first group, the  $d/d_N$  values are smaller and almost constant. Whereas, in the second group,  $d/d_N$  values are larger and vary linearly. The first



**Fig. 14.** Parity plot of the experimental values of  $d/d_N$  and  $d/d_N$  values evaluated by the correlations of aqueous phase drop formation at downward oriented nozzles and organic phase drop formation at upward oriented nozzles. Error bars are  $\pm 7\%$  error bars around experimental data of aqueous dispersed case.

group pertains to larger nozzle diameter (3.04 mm) and the second group pertains to smaller nozzle diameter (1.91 mm). For larger nozzle diameter, nozzle velocities are also small and the Froude number is of the order of  $10^{-2}$  or smaller. Thus, for larger nozzle diameter, the term containing Froude number in the correlation of Eq. (14) becomes almost constant irrespective of the value of Froude number. Therefore, for larger nozzle diameter, the  $d/d_N$  values depend primarily on Eötvös number i.e. on nozzle diameter and physical properties only and not on velocity. Therefore, for larger nozzle diameter,  $d/d_N$  values change only marginally as reflected by the data corresponding to the first group. For smaller nozzle diameter, for which velocities are also larger, the Froude number is at least one order of magnitude more than when the nozzle diameter is larger. Thus, for the case of smaller nozzle diameter, the term containing Froude number in the correlation of Eq. (14) varies with Froude number i.e. with velocity. Thus a change in velocity leads to a change in  $d/d_N$  value which is reflected in the second group of the data. This can also be explained by the force balance as discussed in Section 3.2.1. As the nozzle diameter increases, the contribution of kinetic force ( $F_M$ ) in the force balance equation diminishes whereas the contribution of the interfacial tension force ( $F_i$ ) increases. Thus, for a larger diameter nozzle, the force balance is dictated more by the interfacial tension force which reflects in the velocity or the flow rate of dispersed phase making only limited impact on drop diameter. Thus for larger diameter nozzle,  $d/d_N$  values do not change significantly, as observed in Fig. 14.

#### 4. Conclusions

Experiments are carried out with dilute nitric acid – 30% (v/v) TBP in dodecane system to compare aqueous phase drop formation at downward oriented and organic phase drop formation at upward oriented submerged nozzles. For identical conditions, the drops of the aqueous phase formed at downward oriented nozzles are smaller than the drops of the organic phase formed at upward oriented nozzles. Drop detachment length is more and drop detachment time is less for aqueous phase drop formation at a downward oriented nozzle compared to organic phase drop formation at an upward oriented nozzle. As observed for organic phase drop formation at upward oriented nozzles, drop diameter does not change monotonically with change in dispersed phase

velocity for aqueous phase drop formation at downward oriented nozzles. The drop diameter and drop detachment length increase with increase in nozzle diameter for aqueous phase drop formation at downward oriented nozzles. Drop detachment time reduces with increase in dispersed phase velocity and nozzle diameter for aqueous phase drop formation at downward oriented nozzles. For aqueous phase drop formation at downward oriented nozzles, drop diameter and drop detachment time reduce with an increase in nitric acid concentration. The drop detachment height does not vary monotonically with nitric acid concentration. These trends are the same as that of organic phase drop formation at upward oriented nozzles reported in our previous study [27]. The experimental data on aqueous phase drop formation at downward oriented nozzles are regressed to obtain a correlation for drop diameter. The correlation fits the experimental data with an average absolute relative error of 2.55%. The results and insights presented in this study will be useful for the researchers and practicing engineers working in the field of solvent extraction in nuclear fuel cycle where extraction of precious metals from nitric acid medium using 30% TBP in dodecane is quite common.

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