THE EFFECT OF VISCOSITY ON DROPLET SPLITTING IN ARD AND RDC EXTRACTORS

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It has been found that the size of droplets originated by mixing depends not only on the interfacial tension and the parameters of mixing but also on the viscosity of the droplet and its surroundings. A semi-theoretical relationship has been proposed for the calculation of the size of the droplet. The validity of this relationship has been tested by measurements on ARD and RDC columns 250 mm in diameter and 1 m long.

The hydrodynamic behaviour of agitated and other extractors has been satisfactorily described by the relationship between the hold-up of the dispersed phase, X, and the flow rates U_d and U_c (ref.¹)

$$\frac{U_{\rm d}}{X} + \frac{U_{\rm c}}{1-X} = u_0(1-X)\exp(z-4\cdot 1)X.$$
 (1)

The parameters here are the velocity of single droplets in an infinite extent, u_0 , and the coalescence coefficient, z. This relationship forms basis for the computational procedure of the column diameter. A proven equation for the calculation of drop velocities from their known size in agitated extractors is given below²

$$u_0 = 0.249 d_{43} (g^2 \Delta \varrho^2 |\varrho_c \mu_c)^{1/3} .$$
 (2)

The size of the droplets gives the correlation³

$$We = 45.2 \exp(0.0887 \Delta D) \left(\frac{h_{\rm m}}{D_{\rm K}}\right)^{0.46}$$
(3)

valid for the region of intensive mixing. Then we have

$$d_{43} = 1.055 \left(\frac{\sigma_i}{\Delta \varrho g}\right)^{0.5} \tag{3a}$$

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for the region of moderate mixing and for the transition region the following expression has been found useful

$$We = 2.7 \cdot 10^4 \text{ Re}_{M}^{1.42}$$
 (3b)

From the above three relations the relevant formula is that yielding the least diameter droplet.

The validity and practical utility of this approach has been tested several times^{4,5} for columns or their sections where the hold-up could be regarded constant and with systems whose viscosity did not deviate appreciably from current values.

The study of the behaviour of dispersed viscous liquids has been conducted as it has turned out that such liquids behave anomalously in extractors and, at the same time, a number of industrial applications of liquid extraction works with high viscosity materials, in particular oils. However, nothing has been known about the character of the mentioned deviations. A simple graphical processing of experimental data in Fig. 1 pointed at the following facts: viscosity of droplets has a marked effect on their size; viscosity in all cases increases the size of droplets; viscosity interacts with the rotational speed; experimental data exhibit good reproducibility and their dependence on viscosity and on rotational speed display systematic trends.

Since the size of droplets varies with the viscosity of the d-phase and since neither the present theory nor experimental data recognize such a dependence, it is necessary: correct the theoretical model of droplet splitting in order that it might account for the effect of viscous forces; assume that in the region of low viscosity of the *d*-phase covered so far by experiments this effect was not significant.



FIG. 1 The effect of droplet viscosity on its size

THEORETICAL

The thus far published papers on the size of droplets in agitated vessels mostly did not take into account the effect of viscosity of the dispersed phase on the splitting of the droplets. Theoretical models in particular, did not account for even the effect of viscosity of the continuous phase⁶⁻⁸. Experimentally oriented papers working toward empirical correlations encompass the effect of viscosity of the continuous phase, but it is not apparent to what extent this is due to the fact that these works ignore coalescence and the effect of hold-up on the size of the droplets, and, eventually, the effect of viscosity on the power input of the impeller.

In this respect we shall start from the results of an earlier paper³, supplemented by the effect of viscous forces. The theoretical model shall be based on the following assumptions:

I) The cause for the deformation and consequent splitting of a droplet is the dynamic pressure exerted on the droplet.

II) The dynamic pressure acting on the droplet may be expressed by

$$P_{\rm dyn} = C_1 \varrho_{\rm c} u_{\rm c}^2 , \qquad (4)$$

III) The splitting of the droplet starts by deformation bringing about internal circulation.

IV) For the assessment of the rate of internal circulation it is assumed, in accord with the theoretical solutions^{9,10}, that

$$\frac{u_{\rm d}}{u_{\rm c}} = f\left(\frac{\mu_{\rm c}}{\mu_{\rm d}}\right) \tag{5}$$

and that this unknown function may be sufficiently accurately approximated by an exponential relationship as

$$\frac{u_{\rm d}}{u_{\rm c}} = C_5 \left(\frac{\mu_{\rm c}}{\mu_{\rm d}}\right)^{\gamma}.$$
 (6)

V) The balance of pressures acting on the droplet of the critical (maximum) size must incorporate, apart from the dynamic pressure, (according to assumptions I and II) also the internal (surface) pressure and the result of the internal (viscous) forces in this way:

$$(P_{\rm dyn} = P_{\rm int} + P_{\rm visc})_{\rm crit} \tag{7}$$

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VI) The internal pressure is expressed in accord with the earlier papers³ as

$$P_{\rm int} = C_2 \sigma_{\rm i}/d \;. \tag{8}$$

VII) The internal viscous forces, interpreted as a viscous pressure, are taken to be directly proportional to the velocity of internal circulation, u_d , internal viscosity, μ_d and indirectly proportional to the size of the droplet

$$P_{\rm visc} = C_3 \mu_{\rm d} \, u_{\rm d}/d \, . \tag{9}$$

VIII) In accord with the earlier findings about the tangential velocity of motion of the continuous phase in the agitated compartment we may write

$$u_{\rm c} = \Gamma_{\rm i}(nD) , \qquad (10)$$

where the constant Γ_i characterizes the *i*-th geometry of the agitated compartment and the impeller and it is independent of the rotational speed.

IX) The constants C_1 , C_2 , C_3 , C_5 and γ are independent of the geometry of the compartment and for the RDC and ARD extractors assume an universal value.

X) The character of distribution of the drop sizes is such that 11,12 we may write the proportionality

$$\bar{d}_{pq} \sim d_{\max} \,. \tag{11}$$

A combination of Eqs (4) to (11) yields:

$$\frac{\alpha}{\Gamma_i^2} \frac{1}{We} + \frac{\beta}{\Gamma_i} \frac{1}{Re_d} \left(\frac{\mu_c}{\mu_d} \right)^{\gamma} = 1 , \qquad (12)$$

where

$$We = \bar{d}_{43} n^2 D^2 \varrho_c / \sigma_i = (\bar{d}_{43} / D) . We_M , \qquad (13)$$

$$Re_{d} = \overline{d}_{43}nD\varrho_{c}/\mu_{d} = (\overline{d}_{43}/D)\left(\frac{\mu_{c}}{\mu_{d}}\right)Re_{M}$$
(14)

 α , β , γ are universal constants and such that: α is a positive number, β may generally take both positive and negative values depending on the direction of internal rotation. The applicability of this expression and the effect of individual phenomena from which it has been derived, equally as the magnitude of the constants must be assessed experimentally.

EXPERIMENTAL

The measurements were carried out in a test glass extractor 0.25 m in diameter and 1 m long. The types and geometrical parameters of used internal parts could be varied and have been summarized in Table I. The measurements with viscous liquids were carried out with an ARD column, designated 29. Also incorporated into the processing were all earlier available data³ with other built-ins and other liquids.

The effect of viscosity was tested on the system: Naphta-R-460 oil-trichlorethylene and water. The addition of oil was used to adjust viscosity, trichlorethylene to adjust density of the mixture (Table II). The used mixtures and their properties are reviewed in Table III.

The tests were carried out as follows: A series of runs was performed with a given column, liquid of given properties and a given rotational speed of the impeller, while altering the flow rates of liquids and measuring the hold-up of the dispersed liquid. Fitting the data to the log-log form

Column (designation)	Туре	D _K m	D m	ΔD m	D _i m	h _m m	3
01	RDC	0.25	0.10	0.075	0.15	0.05	0.36
05	RDC	0.25	0.10	0.075	0.18	0.02	0.52
06	RDC	0.25	0.12	0.065	0.15	0.02	0.36
08	RDC	0.25	0.12	0.065	0.15	0.10	0.36
29	ARD	0.25	0.10	0.02		0.02	0.179
30	ARD	0.25	0.12	0.01		0.02	0.179

TABLE I Parameters and designation of used columns

TABLE II Review of physical properties of used systems

Continuous phase	Dispersed phase	$\frac{\varrho_{\rm c}}{{\rm kg/m}^3}$	$\frac{\varrho_d}{kg/m^3}$	μ _c mPa s	μ _d mPa s	σ _i mN m
Water	oil 10	994·0	888.3	1.030	7.68	30.11
Water	oil 20	1 001.4	886.4	1.1018	17.05	29.04
Water	oil 40	1 000.0	886.0	1.050	36.77	31.84
Water	oil 70	1 000.0	882.0	1.048	61.56	33.36
Water	oil 100	1 000.0	891.0	1.024	90.44	30.10
Water	oil 120	1 000.0	890.3	1.072	108.24	32.40
Water	oil 140	989·0	890.0	1.023	124.65	31.75

of Eq. (1) the magnitude of the velocity u_0 was obtained, which in turn served to compute the magnitude of d_{43} from Eq. (2) (the justification of this mean value is given elsewhere²). The agreement of the measured hold-ups with Eq. (1) and the method of fitting is illustrated in Fig. 2. Values free of the effect of viscosity for comparison were computed from Eq. (3). The description of the experimental set-up, its scheme, as well as a more detailed procedure is given in the earlier paper¹.

The experimental conditions and the results of tests were characterized by dimensionless quantities according to these expressions:

$$Re_{\rm Md} = nD^2 \varrho_{\rm c}/\mu_{\rm d} \tag{15}$$

$$We_{\rm M} = nD^3 \varrho_{\rm c} / \sigma_{\rm i} \tag{16}$$

$$Vi = (\mu_c/\mu_d)^{1.17}$$
(17)

and the relationships given by Eqs (14) and (13). The results are given in Table IV.

The evaluation of Eq. (12) was carried out in several steps in order to verify its individual parts.

RESULTS

First we have verified additivity of the surface and viscous forces using the set of data with column 29 which were most accurate and most numerous. Fig. 3 plots the dependence 1/We versus $1/Re_d$, which seems to well comport the presumed linear course. The intersection of the fitted straight lines falls clearly off the zero point, into the region of positive 1/We. The slope of these straight lines is, according to the assumption IV, a function of viscosities tg $\varphi \sim (\mu_c/\mu_d)$, which is according to Fig. 4

TABLE III Physical properties of test systems (20°C)

System						
continuous phase	dispersed phase	- e _d kg/m ³	$\frac{\rho_{c}}{kg/m^{3}}$	μ _d mPa s	e _c mPa s	σ _i mN m)
Water	TOL	871.6	998-2	0.582	1.004	24.48
Water	BAC	881.5	997.3	0.717	1.004	12.28
Water	AAL	829-2	993-9	4.120	1.101	7.50
Water	PF	800.0	1 000.0	1.440	1.002	37.00
Water	TRI	1 464.4	998.5	0.542	0.926	37.40
Water	EE	986-5	708·0	0.216	1.280	12.00
Water	PE	645.5	1 000.0	0.271	1.000	48.00

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TABLE IV

Results of measurements

We	Re	Re/Vi	We	Re	Re/Vi		
	Column 01			Column 29			
18-6	2 363	1 250	41.1	17.8	2 089		
36.3	3 067	1 622	46.5	21.6	2 535		
40.5	2 910	1 539	120.6	48.2	9112		
29.9	1 892	1 000	102.6	37.1	7 013		
30.9	1 736	920	81.7	27.2	5 142		
35.6	1 428	755	77.4	23.8	4 500		
11.3	83	390	62.1	17.7	3 346		
19.4	106	500	54.6	14.6	2 760		
10-1	706	470	52.0	13-0	2 457		
18-2	957	630	50.8	12.4	2 344		
27.6	1 1 2 2	2 175	113-5	37.0	8 186		
46.8	1 806	2 761	92.9	27.8	6 1 5 0		
45-8	1 284	1 963	76-1	21.0	4 646		
25-1	645	986	1				
			i	Column 30			
	Column 05						
			40.0	4 270	2 250		
32.8	1 014	1 544	40.1	3 380	1 781		
30.9	866	1 324	46.0	3 280	1 730		
18-2	469	717	45.9	10 200	2 220		
			36-0	7 011	1 530		
	Column 06		37-0	6 514	1 420		
36.6	2 295	1 230	59-2	9 602	2 091		
40.1	2 308	1 230	42.1	2 900	363		
38-5	1 650	2 530	49.3	2 1 0 9	3 250		
11.3	484	2 550	47.0	1 750	2 700		
9.7	417	640	60.0	1 930	2 970		
21	417	040	57-1	1 642	2 522		
	Column 08		46-0	1 180	1 820		
73.0	2 343	3 600		Column 29			
57-1	1 835	2 823	1				
52.0	1 681	2 580	59-5	262	2 746		
35-9	1 156	1 780	48.8	194	2 034		
			27.6	101	1 059		
	Column 29		19-1	64.4	675		
	A A 7		48.9	99.9	2 448		
56.6	30.7	3 603	56.7	105-4	2 583		
40.3	20.2	2 371	46.1	78.5	1 924		
37.5	17•4	2 042	27.1	42.6	1 044		

We	Re	Re/Vi	We	Re	Re/Vi
	Column 29			Column 29	
19.6	28.6	701	57-4	12.1	2 677
68.4	51.0	3 269	117-4	32.6	8 980
71.3	67.4	4 321	108-8	27.7	7 630
59.5	51.6	3 308	90.4	21.3	5 868
42.4	33-9	2 173	80.2	17.5	4 821
39.4	29.2	1 872	74.9	15-3	4 215
33-9	23.5	1 506	69.7	13-4	3 691
63.7	41.4	4 859	58-0	10.4	2 865
64-2	37.9	4 448	72.6	4 925	2 605
76.3	19.6	4 3 3 6	56.5	2 858	1 512
59-3	14-2	3 142	43.0	188	883
36.7	8-2	1 814	39.7	1 191	783

in good agreement with the reality and satisfactory for a value $\gamma = 1.17$. This value was used in turn in the regression of the data of *We versus* $Re_d[(\mu_c/\mu_d)^{1.17}$. This fitting yielded for the data from column 29 and 30 (ARD columns) constants Eq. (12):





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TABLE IV

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$$\alpha / \Gamma_{i}^{2} = 8 \ 073 \cdot 4 = B$$

$$\beta / \Gamma_{i} = -222 \cdot 3 = A . \tag{18}$$

Fig. 5 is a confrontation of the computed course with the experimental data.

The same figure shows also data points from measurements on columns of other types. It turns out that these data are also sufficiently well matched by the proposed



FIG. 3 The relationship between We and Re_d





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constants (18). Their scatter, however, owing to their lower accuracy, is somewhat higher and does not allow evaluation of eventual, small deviations in the constants Γ_i . Until there are more accurate data from columns of other geometries available, the constants A and B (Eq. (18)) shall be taken not to be versatile for the RDC and ARD columns.

DISCUSSION

In connection with the newly found expression for the size of the droplets it is necessary to discuss the reasons which lead to the earlier (3) expression according to which for a given column:

$$We = \text{const.}$$
 (19)

The magnitude of the constant here depended solely on the column geometry. An earlier paper suggests³ that the validity of Eq (19) was confined to the region of developed turbulence and an empirical relation, incorporating the Reynolds number and hereby also viscosity, was recommended for the transition region. Indeed, according to the new equation (12) the effect of viscosity diminishes also with the increasing Reynolds number which ultimately may lead to its neglection. The new relation (12) correlates satisfactorily the data also from the transition region. Another leading to the neglection of the viscosity effect provides the following reasoning:

The dependence of We on Re, shown in Fig. 5, may be in log-log coordinates approximated by a straight line

$$We = a(Re/(\mu_c/\mu_d)^{\gamma})^{\mathbf{b}}.$$
⁽²⁰⁾





For b = 0.7, which is an approximate value found in the operating region, we obtain

$$Re \sim \left(\frac{\sigma\mu_{\rm c}}{D}\right)^{3.3} \left(\frac{\mu_{\rm c}}{\mu_{\rm d}}\right)^{2.3\gamma}$$
 (21)

Hence $Re \neq f(n \cdot D)$ and for the tests with a single liquid system and a single geometrical configuration we also find We = const.

The effect of physical properties probably canceled out in the experimental program. For instance, the system butanol-water and amylalcohol-water, which in the experimental program represented low interfacial tension systems, display also high viscosity and the systems with a low interfacial tension do not differ mutually neither in the interfacial tension nor viscosity.

On the contrary, at high values of Re, b increases and Eq. (21) does not hold even approximately; in the transition region the effect of Re must be considered, as, in fact, has been found also experimentally.

Independently of these facts, the correlation of the type of Eq. (19) may be compared with the correlations as in Eq. (12) on the set of data with ARD columns (columns 29 and 30) and with all columns. This shall be made by the analysis of variynce. From the results it is apparent that Eq. (12) provides results with a smaller variance (by about 50%) and that the significance of its introduction is conspicious ($P \le 0.01\%$). The unit relative standard deviation of the correlation (12) decreases, compared to the correlation (19) by 35%, while the experimental errors are for both sets approximately equal.

In view of the method of evaluation of the size of the droplets from column holdups, the standard deviation may be regarded to be the experimental error. The accuracy of the average is $\pm 3\%$.

Since the measurements of the type employed in this work are not sufficient for the identification of the effect of geometrical parameters Γ_i on the velocity u_e , it would be proper to direct further research on their independent investigation.

So far unclarified remains the effect of the unsteadiness of the size of the droplets. It must be assumed that more correct and more accurate results would be obtained by measuring on columns substantially longer than 1 m. This as well should be the subject of further research.

CONCLUSION

A semi-theoretical model has been developed of the droplet splitting in ARD and RDC extractors taking into account the effect of viscosity of both liquid phases. The model has been confronted with extensive experimental data and it has been found that it fits substantially better than earlier papers³. The parameters of the model

have been evaluated and it has been found that as a first approximation the parameters may be taken to be independent of the geometry of the extractor. This has a considerable advantage for the modelling of extractors – for the scale-up to industrial size since no risky extrapolations are needed. Eq. (12) may be used with a high confidence not only for the calculation of extractors working with highly viscous liquids but also for a significant improvement of precision on the region of current viscosities.

From the results of this work it may be concluded that also the theoretical models of droplets splitting (Kolmogorov⁶ and others, *e.g.*, refs^{7,8}) are oversimplified.

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LIST OF SYMBOLS

D	impeller diameter (m)
ΔD	distance of impeller from wall (m)
Dĸ	column diameter (m)
υ	superfacial velocity of liquid phase in column $(m s^{-1})$
Х	liquid hold-up
P_{dyn}	dynamic pressure (Pa)
Pint	internal pressure (Pa)
Pvisc	result of internal viscous forces (Pa)
Red	Reynolds number related to droplet and dispersed phase
We	Weber number
ReM	Reynolds number for mixing
WeM	Weber number for mixing
ſ	general function
a	constant of a plane
b	constant of a plane
dpg	mean droplet diameter (m)
d	droplet diameter (m)
g	acceleration due to gravity $(m s^{-2})$
u _c	velocity of continuous phase (m s ⁻¹)
u _d	internal velocity of dispersed phase (m s ⁻¹
u ₀	characteristic velocity of droplet (m s ⁻¹)
n	frequency of revolution of the impeller (s^{-1})
α	constant, Eq. (8)
β	constant, Eq. (8)
γ	exponent, Eq. (8)
Γ_{i}	constant, Eq. (8)
З	porosity
Qc	continuous phase density (kg m ⁻³)
<i>Q</i> _d	dispersed phase density $(kg m^{-3})$
μ_{c}	continuous phase viscosity (Pa s)
μ_{d}	dispersed phase density (Pa s)
σ_i	interfacial tension $(kg s^{-2})$
φ	angle
Δ	deviation

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