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CHEMICAL PROCESS SYSTEMS LABORATORY

Analysis of Drop Size Distributions Produced in Dilute, Agitated Liquid-Liquid Systems

by

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RESEARCH REPORT

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reasoned that very viscous drops break either by stretching into dumbell-like structures or by stripping of small drops from a parent. In the former case, fracture yields a few large daughters and many satellites. For the latter the final distribution consists of many small droplets and a few large drops which have become too small to undergo further stripping. In either case, the equilibrium distribution consists of two types of drops, satellites and surviving daughters. In the language of atmospheric physics, the measured distribution can be viewed as resulting from the combination of two sources. In order to interpret the data in a meaningful manner, it is useful to extract the parameters which describe the individual distributions and to subject these to further analysis.

The purpose of this work is to develop a rational procedure for inverting the measured distribution and to demonstrate its validity to the aforementioned data. A procedure is described by which the condition number is employed as the criterion for determining which moments yield the most accurate estimates of the individual distribution parameters when the measured distribution is described as the sum of two log-normal distributions. The procedure is tested using contrived data and then applied to the stirred tank data. The procedure is shown to be generally valid but requires some modification to account for known errors in data acquisition. The results in terms of individual source mean, variance and number fraction are discussed by reference to relevant stirred tank physical phenomena.

Approach

The analysis of the size distribution depends upon three assumptions:

1. The measured distribution can be described as the sum of two log-normal distributions. Examination of the high viscosity data presented by Calabrese et.al.(1986) show this to be a reasonable assumption. Furthermore, use of this functional form facilities analysis since all moments can be expressed in a closed analytical form.

2. The parameters of the individual distributions can be obtained by matching moments of the distribution function with those calculated from the experimental measurements.

3. The criteria for selecting the set of six moments are the minimum condition number and the value of the largest and/or smallest moment.

Implicit in the analysis is the assumption that the experimental data consist of measurements of individual drop size. The principal errors arises from the fact that the sample may not be representative of the actual drop population. This is especially likely for the largest drops which control the largest positive moments and which are comparatively rare in the distribution. This may also be the case for the smallest drops, which are larger in number and control large negative moments, if these are beyond the resolution of the measurement technique.

Condition Numbers

Condition numbers are quantative criteria which characterize the magnitude of perturbations in the solution vector of a set of linear algebraic equations due to fluctuations in the forcing function vector. They were originally used to analyse the effect of roundoff error in numerical computation but have recently been applied to experimental design and a priori evaluation of data inversion algorithms. Yu (1983) tested condition numbers as a means of characterizing the performance of several aerosol classifiers in the determination of size distribution. Farzanah et.al. (1985) employed condition number criteria to determine the optimal set of chemical indicators elements for the process of associating atmospheric aerosols with their source. Kaplan et.al. (1985) developed a procedure for a priori evaluation of power law correlations widely used in engineering.

For application to this study, it is assumed that the problem of inverting multi-source size distribution data can be characterized by a simultaneous set of linear algebraic equations

$$[\,A\,]\;[\,Y\,] = [\,B\,] \qquad (1)$$

Vector B is a forcing function and represents measured quantities or the difference between measured and estimated

$$M_p = \sum_{i=1}^{2} G_i(P) + \sum_{i=1}^{2} G_i(P)\left[\frac{\Delta f_i}{f_i} + P\frac{\Delta X_i}{X_i} + P^2\sigma_i\,\Delta\sigma_i\right] \tag{8}$$

The first term on the right hand side of Eq.8 represents the estimated value of the moment while the seond term, which ideally approaches zero, represents the perturbation. The minimum condition number tells us the particular values of P for which this term will be a minimum, independent of the uncertainty in the data.

Let P_J where J=1 to 6 be the set of moments (including P=0) under consideration. To obtain the particular form of Eq. 2 employed here, let the components of the solution vector Y_k, where k=1 to 6 be the $\Delta f_i/f_i$, $\Delta X_i/X_i$ and $\sigma_i\,\Delta\sigma_i$ where i=1 then 2. Then the ideal solution vector has all components equal to zero and the coefficients of the A matrix are $G_i(P_J)$, $P_J G_i(P_J)$ and $P_J^2 G_i(P_J)$, respectively. The algebraic equation has the form

$$A_{kJ}\; Y_k = B_J \tag{9}$$

where the components of the forcing function vector are

$$B_J = M_{pJ} - \sum_{i=1}^{2} G_i(P_J) \tag{10}$$

Eq.10 states that the components of the forcing function are equal to the true value of the moments minus the estimated value.

In computing the $q = \infty$ condition number from Eq.3, each row of the A matrix is normalized as previously discussed. It is then seen that the value of C_∞ depends only upon the values of P and the ratio G_2/G_1. This ratio can be calculated from a priori estimates of the f_i, X_i and σ_i to determine the six particular values of P that yield the lowest condition number. In this way we can choose, a priori, the most suitable set of moment equations for accurate inversion of the drop size distribution data.

Choice of Moment Equations

Rather than scrutinize each drop size data set individually, we employ a less demanding scheme to determine if a particular choice of moments is generally more suitable

than others. We note that positive moments are controlled by the larger drops while negative moments emphasize the smaller drops. Three test cases are chosen as shown in Table 1. Case 1 equally weights the distribution since both positive and negative moments are employed and the maximum absolute value of P is not too large. Case 2 gives higher weight to the larger drops but does not emphasize the largest drops. Case 3 contains large positive moments and therefore heavily weights the largest drops. These cases are tested using ficticious data sets that contain no error; that is, values of the f_i, X_i and σ_i are selected and a contrived combined distribution representing measured data is calculated directly from them. This distribution is used in the inversion algorithm to extract the already known individual distribution parameters.

The first test is based upon the individual distributions having the same standard deviation; that is $\sigma_1 = \sigma_2 = 0.3$. Then, according to Eqs.5 and 7 the ratio G_2/G_1 depends only upon $X_R = X_2/X_1$ and $f_2 = 1-f_1$. Figure 1 shows a plot of C_∞ versus f_2 for $X_R = 2$ which represents individual distributions of similar means. Case 1 always has the smallest condition numbers with Case 2 being the least suitable. Similar computations for $X_R = 10$ (widely separated distributions) show that Case 1 always has the minimum C_∞ for $f_2 > 0.002$. Case 3 is more suitable for smaller f_2. For all cases, $X_R = 10$ has smaller condition numbers than $X_R = 2$ indicating that widely separated means are easier to analyze.

Table 2 shows the effect of standard deviation on C_∞ for $X_R = 10$ and 3 different values of f_2. Condition numbers are reported for Case 1 and ratios of condition numbers are reported for the other cases. Again Case 1 is nearly always better except for very small f_2. Similar results were obtained for $X_R = 2$.

Figure 2 shows the effect of sample size on the retrieved parameters for $\sigma_1 = \sigma_2 = 0.6$, $X_R = 6$ and $f_2 = 0.9$. For each sample size, ten times as many drops comprised the test distribution. Ten trials were run with one-tenth

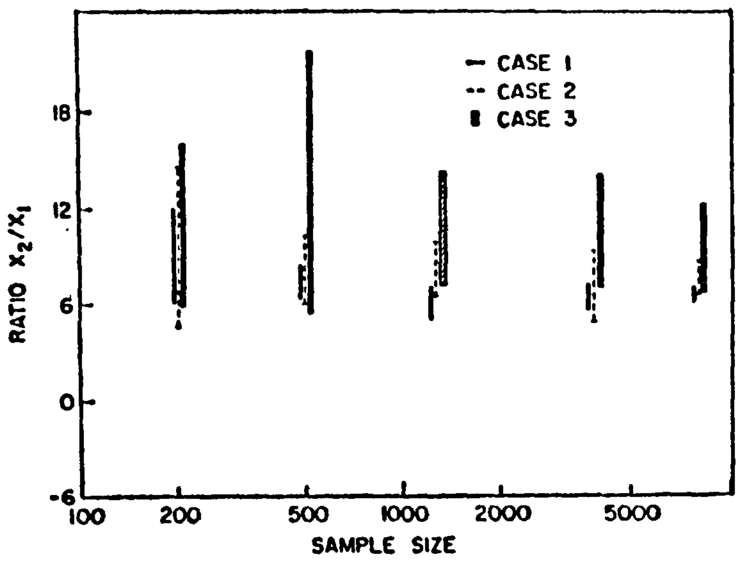

Figure 2. Effect of Sample Size on Retrieved Value of X_R for $\sigma_1 = 0.6$ and $f_2 = 0.9$. True Value is $X_R=6$.

The experimental details are given by Calabrese et.al. (1986). What must be noted is that for the oils considered here, drop sizes ranged from tens to thousands of microns. These were measured manually from the photographs and binned into 50 or 100 µm intervals. A significant number were smaller than 50 µm, the resolution of the photographic technique. The authors placed these in the 50 µm bin and circumvented the problem of inaccuracy in the small size tail be reporting cumulative frequency. Here we expect significant error in calculation of large negative moments.

Table 3
Experiments and Sauter Mean Diameter

Run	Tank Diameter m	Impeller Speed RPS	Sauter Mean Diameter, µm Experiment	Case 1	Case 4
Dispersed Phase Viscosity, μ_d=4.43 Pa.s					
1	0.142	2.45	2781	2781	2848
2		3.05	2442	2440	2546
3		3.27	2263	2264	2333
4	0.213	1.83	2965	2965	3128
5		2.00	2723	2723	2899
6		2.20	2427	2427	2515
7		2.42	2231	2230	2287
8		2.67	2131	2131	2223
9		2.85	2087	2087	2209
10		3.15	1786	1752	1811
11	0.312	2.10	2095	2095	2205
12		2.38	1827	1827	1920
13	0.391	1.70	2136	2136	2303
14		2.00	1977	1968	2057
Dispersed Phase Viscosity, μ_d=10.5 Pa.s					
15	0.142	3.20	3206	3206	3254
16		3.50	2906	2906	
17		3.95	2851	2851	2888
18		4.67	2357	2357	2410
19	0.213	2.05	3536	3536	3624
20		2.35	3261	3261	3291
21		2.72	2640	2640	2679
22	0.312	1.40	3976	3976	4157
23		1.60	3836	3836	4022
24		1.75	3365	3365	3474
25		1.90	3066	3083	3245
26		2.05	2830	2830	3065
27		2.20	2765	2765	2856
28	0.391	1.50	3305	3309	3602
29		1.90	2721	2721	2823
30		2.15	2637	2637	2838

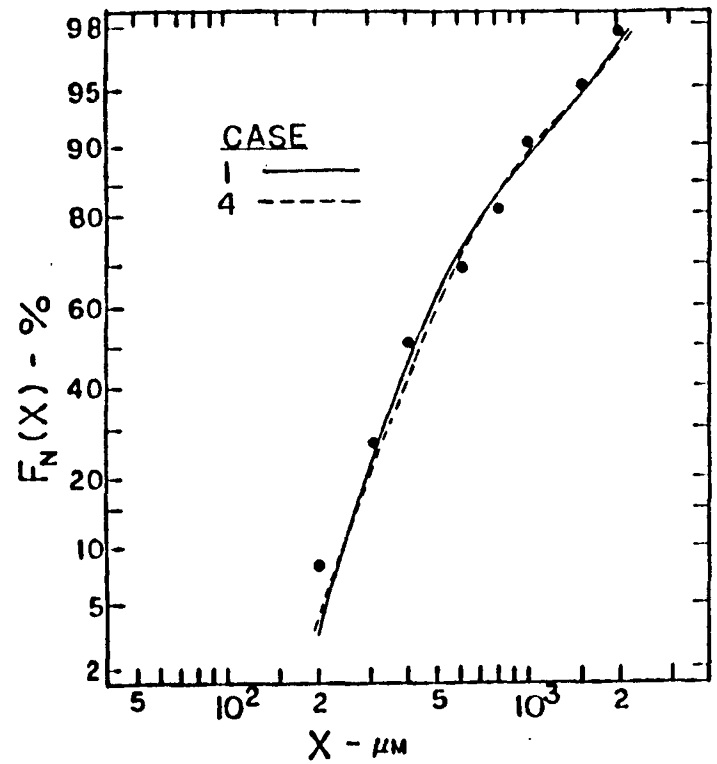

Figure 3. Comparison of Predicted and Measured Cumulative Number Frequency for Run 3; μ_d = 4.43 Pa·s; T = 0.142m; N = 3.27 RPS.

<u>Discussion</u>

In turbulent, stirred tank contactors, Sauter mean diameter increases with increasing dispersed phase viscosity and tank size and with decreasing impeller speed. The same trends can be seen from Tables 3 and 4 for X_2, the mean daughter drop size. However, the mean satellite size, X_1, does not appear to display any consistent trend with system variables. The number fraction of satellite droplets, f_1, is larger for the more viscous oil. This may be related to the fact that a more viscous globule can deform to a greater extent before fracture, thereby increasing the potential for satellite production. The

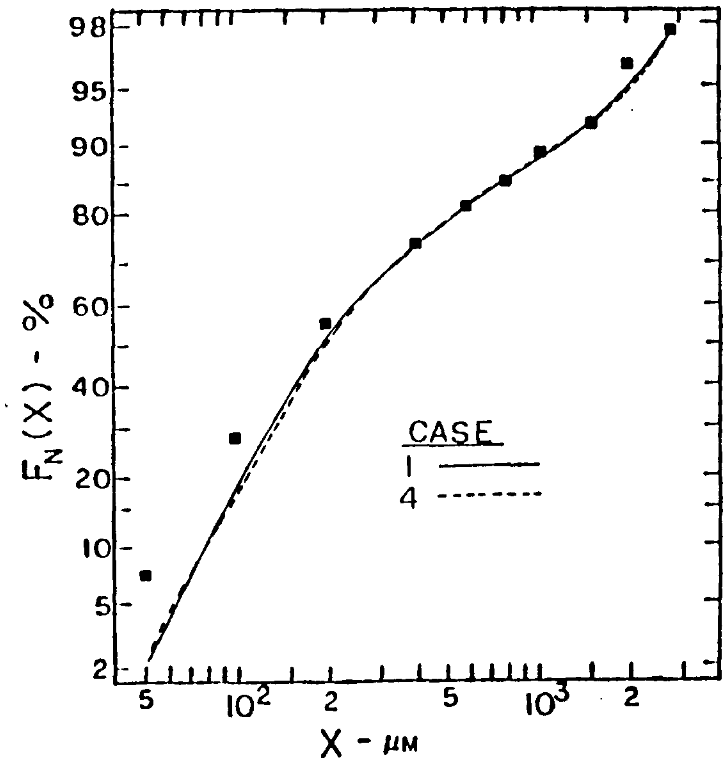

Figure 4. Comparison of Predicted and Measured Cumulative Number Frequency for Run 22; μ_d = 10.5 Pa·s; T = 0.312m; N = 1.40 RPS.

results also show that the standard deviation of the satellite distribution increases with viscosity but that for the daughters appears independent of viscosity. However, σ_2 does increase with tank size.

Now that the individual distribution parameters have been acquired, they can be subjected to interpretation based upon mechanistic arguments of the type commonly employed for less viscous dispersed phases (e.g. see Talvarides and Stamatoudis, 1981). This will be the subject of future work.