SOLUTION OF INVERSE PROBLEMS IN POPULATION BALANCES—II. PARTICLE BREAK-UP

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Abstract—A mathematical and computational procedure called the inverse problem is developed to extract quantitative information from transient particle size distribution measurements. The population balance framework describes the evolution of these transient size distributions. This paper describes an inverse problem for the determination of the size specific breakage rates and daughter drop distributions from transient size distribution measurements when these distributions evolve to a “self-preserving” or similarity distribution. The experimental time scaled with respect to the timescale of breakage is used as the similarity variable.

A test for the existence of similarity is developed. This test uses only the available transient size distribution data. The result of this test is also used to determine breakage rate information. The determination of the daughter drop distribution is an ill-posed problem. The ill-posedness is overcome by using the property that the distribution is a monotone function. Analysis shows that the asymptotic behavior of the daughter drop distribution can be determined from the experimental similarity distribution. Incorporating this additional information into the solution strategy has resulted in significantly improved solutions of the inverse problem. The optimum solution is chosen such that the similarity distribution predicted using this solution has error of the same order of magnitude as the error in the experimental similarity distribution. Several examples of the inverse problem are outlined.

1. INTRODUCTION

Dispersed phase systems find important uses in a wide range of chemical engineering applications. For example, liquid–liquid separations and dispersed phase reactions are processes where the enhanced surface area created by dispersing one phase in another is utilized to great advantage. The mechanism used to create the dispersion, e.g. mixing, causes particles of the dispersed phase to break and coalesce and thereby creates a distribution of particle sizes. This particle size distribution affects the overall performance of the transport process occurring in the system. The evolution of this size distribution can be written in terms of the population balance equation. However, we need quantitative information about the particle break-up and coalescence processes to use the equation to predict and control this size distribution.

Inverse problems provide us with a powerful tool to obtain this quantitative information from transient size distribution measurements. In a previous paper (Wright and Ramkrishna, 1992), we have dealt with the solution of the inverse problem for coalescence. The techniques presented in that paper allow us to determine particle coalescence rates from transient size distributions. This paper deals with the solution of the inverse problem for breakage. Our particular interest is in the breakage of drops in stirred liquid–liquid distributions. The inverse problem approach allows us to determine the breakage rate and the daughter drop distribution, i.e. the size distribution of drops formed from the breakage of a larger drop, from transient size distribution measurements.

Since drop breakage is important, many investigators have studied it. The most common approach to study drop breakage has been to model the breakage phenomenon in a stirred environment. However, due to the inherent difficulty of modeling in a turbulent flow situation, many simplifying assumptions are made in the models. Examples of drop breakage models can be found in the works of Coulaloglou and Tavlarides (1977), Narsimhan et al (1979), and Nambiar et al (1992).

Another approach used to study drop breakage has been to directly observe breakage events. Konno et al (1983) used high-speed photography to observe drop breakage. The major drawback of this approach is that it is almost impossible to measure a sufficient number of breakage events. Konno et al measured less than one hundred breakage events.

Our inverse problem technique uses the concept of self-similar size distributions to determine the breakage rate and daughter drop distribution. It...
takes advantage of the constraints that the self-similar distributions place on these functions.

In this paper, we introduce the concept of self-similarity and the choice of the similarity variable. We outline how to test the experimental data for similarity and how that test leads us to the determination of the breakage rate. We describe the computational methods used to calculate the breakage rate and the daughter drop distribution from the resulting similarity distribution. Several examples of the inverse problem technique are given to illustrate the procedure.

2. SIMILARITY AND THE INVERSE PROBLEM

The population balance framework provides us a convenient method to track evolving drop size distributions. For a batch dispersed phase system evolving by pure breakage, the population balance equation can be written in terms of the cumulative volume fraction as:

$$\frac{\partial F(v,t)}{\partial t} = \int_v^\infty \Gamma(v')G(v,v')\partial_v F(v',t)$$  \hspace{1cm} (1)

where $F(v,t)$ is the cumulative volume fraction of drops of size less than or equal to $v$ at time $t$, $\Gamma(v)$ is the breakage rate of drops of size $v$ and $G(v,v')$ is the cumulative volume fraction of drops of size less than or equal to $v$ formed from the breakage of a drop of size $v'$. Our inverse problem then is to determine $\Gamma(v)$ and $G(v,v')$ from experimental $F(v,t)$ measurements.

2.1. Similarity transformation of the population balance equation

Filippov (1962) and Ramkrishna (1974) have shown that under conditions of a power law breakage rate, i.e. $\Gamma(v) = kv^n$, and similar breakage, i.e. $G(v,v') = g(v/v')$, equation (1) admits a similarity transformation of the form $z = v^n t$. Ramkrishna (1974) formulated an inverse problem which took advantage of the conditions under which this similarity was valid to obtain the power law exponent $n$.

A technique using the moments of the similarity distribution was also formulated to determine the daughter drop distribution from the experimental $F(v,t)$ measurements. Several examples of the inverse problem technique are given to illustrate the procedure.

The form given by equation (2) allows larger drops to break more thoroughly than smaller drops, as explained by Narsimhan et al. (1980). It also admits the similarity transformation $F(v,t) \rightarrow f(\xi)$, where $\xi = \Gamma(v)t$. It is significant to note that the above similarity form is not constrained to power law breakage rates. Hence, nonpower law breakage rates can show self-similar behavior if the assumption of similar breakage, equation (2), is valid. Narsimhan et al. (1980) also developed a test to determine if the experimental data showed the above similarity. If the test for similarity was positive, this result could also be used to determine the breakage rate up to a multiplicative constant. They also developed a technique using moments of the similarity distribution, similar to that of Ramkrishna (1974), to determine the daughter drop distribution. They tested their inverse problem technique on experimental data and were able to calculate the breakage rate. Because of difficulties experienced with using the moments technique to determine the daughter drop distribution, Narsimhan et al. (1984) attempted to fit a beta distribution to the $G[\Gamma(v)/\Gamma(v')]$ function. Using this approach and from the test for similarity, they were able to determine breakage rate and the daughter drop distribution for a variety of different liquid-liquid systems.

Wright and Ramkrishna (1992) have very successfully determined aggregation rates by using a similarity transformation of the form $z = v/S(t)$, where $S(t)$ is an average volume of the distribution. In our investigations, we considered using this type of transformation for the analysis of breakage. An analysis of this transformation shows that it is only valid when $\Gamma(v)$ is a power-law function. As mentioned earlier, the transformation $\xi = \Gamma(v)t$ is not restricted to power-law breakage functions. Since the $\xi$ transformation covers a broader range of functions than the $z$ transformation, we have developed the inverse problem for breakage in terms of the $\xi$ transformation.

Substituting the $\xi$ transformation into equation (1) yields:

$$\xi' f'(\xi) = \int_\xi^\infty g'\left(\frac{\xi}{\xi'}\right) \xi' f'(\xi')d\xi'. \hspace{1cm} (3)$$

2.2. Inverse problem formulation

In the similarity transformation $\xi = \Gamma(v)t$, the breakage rate $\Gamma(v)$ is unknown. Unlike the coalescence case, where the scaling volume is an average volume of the distribution and can be easily determined from the experimental data, testing to see if the data follows the above similarity transformation is a non-trivial matter. Narsimhan et al. (1980) have
developed a procedure to test the experimental data for similarity.

Following Narsimhan et al. (1980), if the similarity transformation is valid, for a fixed value of \( F(v, t) \), \( v \) and \( t \) would be related by \( \Gamma(v) = \text{constant} \), since the similarity variable would be constant at constant \( F \). Differentiating this expression with respect to \( v \), we get:

\[
\frac{d\Gamma(v)}{dv} + \left( \frac{\partial t}{\partial v} \right) \Gamma(v) = 0.
\] (4)

Multiplying equation (4) by \( v \) and rearranging, we get:

\[
\frac{d \ln \Gamma(v)}{d \ln v} = \left( \frac{\partial \ln t}{\partial \ln v} \right). \tag{5}
\]

Equation (5) provides the test of the similarity hypothesis. Since the left-hand side of the equation is independent of \( F \), so should the right-hand side of the equation. This means that if we plot \( \ln t \) versus \( \ln\ln v \) for various constant values of \( F(v, t) \) from the experimental data, the slopes of these various curves at any value of \( v \) should be independent of \( F \). Such independence of the slopes must imply that a vertical translation of the various curves must result in their collapse into a single curve. The similarity hypothesis may be deemed to be valid if the foregoing test for collapse of the \( \ln t \) versus \ln \( v \) curves for different \( F \) occurs.

The above technique provides us an appropriate test of the similarity hypothesis. In addition, if the experimental data does show similarity, the collapsed \( \ln t \) versus \( \ln v \) curve provides us additional information on the rate of breakage. From the collapsed curve, \( \partial \ln t / \partial \ln v \) can be estimated at various values of \( v \). Integrating equation (5) from some reference volume \( v_0 \) to \( v \), we get:

\[
\Gamma(v) = \gamma \exp \left[ -\int_{\ln v_0}^{\ln v} \left( \frac{\partial \ln t}{\partial \ln v} \right) \, d \ln v \right]. \tag{6}
\]

where \( \gamma \) is the (unknown) breakage rate of a drop of volume \( v_0 \). At this point, our inverse problem formulation has determined the breakage rate up to a multiplicative constant \( \gamma \). We now need to determine the daughter drop distribution and the constant \( \gamma \).

By redefining the similarity variable as \( \zeta = (\Gamma(v)/\gamma) t \), the population balance equation in terms of the new \( \zeta \) becomes:

\[
\zeta''(\zeta) = \gamma \int_{\frac{v}{u}}^{v} \frac{1}{u} F \left( \frac{\zeta}{u} \right) g(u) \, du. \tag{7}
\]

Equation (7) is the basis of our efforts to determine the function \( g \) and the constant \( \gamma \).

3. COMPUTATIONAL DETAILS OF THE INVERSE PROBLEM

As discussed in the previous section, the inverse problem for breakage can be divided into three parts—testing the similarity hypothesis, determination of the breakage rate if the similarity hypothesis is valid and determination of the daughter drop distribution from the resulting similarity distribution. We will detail each of these steps in this section. The techniques discussed in this section have been tested out on simulated as well as experimental data as will be shown in the sequel. We will first describe how to treat the transient size distribution data to determine if the similarity hypothesis is valid. We will then use the resulting information to calculate the breakage rate function and determine the self-similar distribution. This distribution is the input needed to calculate the daughter drop distribution. We discuss the choice of basis functions used to describe this function and the type of solution technique needed to solve equation (7) to calculate the function.

3.1. Test for similarity

If the transient size distributions show similarity with respect to \( \zeta \), the slope of the \( \ln t \) vs \( \ln v \) curves must be independent of the cumulative volume fraction \( F \). Hence, the \( \ln t \) vs \( \ln v \) curves for different values of \( F \) must all collapse into a single curve if similarity exists.

Previous efforts to determine if the curves all collapse into one (Narsimhan et al., 1980, 1984; Brown and Glatz, 1987) involved translating the different curves in the vertical direction (i.e., parallel to the \( \ln t \) axis). If, by suitable translation, the curves for different \( F \) values collapsed into one, it was determined that the data exhibits similarity. This translation technique involved choosing a reference curve into which all the other curves would be translated and choosing a reference drop volume for the translation. However, most of the curves do not extend up to the reference volume and there is negligible overlap between the different curves which causes problems in determining the translation distance. Considering these drawbacks, we describe a new technique to test the data in the form of \( \ln t \) vs \( \ln v \) curves for the existence of similarity.

Any regular, plane curve, \( y = y(x) \), is uniquely determined if we know the curvature \( \kappa \) as a function of the arc length \( s \) (or of \( x \) ), and the arc length of the curve as a function of \( x \). According to equation (5), if similarity exists, the various \( \ln t \) vs \( \ln v \) curves are all part of the same curve, except that the various parts of that common curve are displaced vertically.
from one another. Hence, if the similarity hypothesis is valid, the arc lengths and curvatures of the different \( \ln t \) vs \( \ln v \) curves should be equal in the sections of the \( \ln v \) axis where the curves overlap. Our test for the existence of similarity then consists of testing if the \( \kappa(x) \) and \( s(x) \) values of the different \( \ln t \) vs \( \ln v \) curves are the same at all \( x = \ln v \) values.

Typically, experimental drop size distribution data is collected at a few (5–10) discrete times. Calculating the arc length and curvature of the curves from such sparse experimental data can lead to large numerical errors. Note that for a curve \( y = y(x) \), the curvature and arc length are given by:

\[
\kappa(x) = \frac{d^2y}{dx^2} \left[ 1 + \left( \frac{dy}{dx} \right)^2 \right]^{1/2} \tag{8}
\]

and

\[
s(x) = \int_{t_0}^t \left[ 1 + \left( \frac{dy}{dx} \right)^2 \right]^{1/2} \, dx. \tag{9}
\]

Hence, curvature and arc length calculations involve determination of first and second derivatives of the curves from experimental data.

In order to circumvent the numerical difficulties associated with calculating derivatives from sparse, noisy experimental data, we have chosen to fit the different \( \ln t \) vs \( \ln v \) curves to a polynomial functional form before calculating the derivatives required for the curvature and arc length. In the various cases we have analyzed with both simulated and experimental data, it was found that the curves could be adequately fitted with a quadratic curve. By fitting the data, we smooth out the experimental error and also obtain a continuous curve to calculate \( \kappa \) and \( s \). The calculated \( \kappa \) and \( s \) values for the different \( \ln t \) vs \( \ln v \) curves are then compared to see if they are approximately equal for all values of \( \ln v \).

3.2. Calculation of the breakage rate

If the similarity hypothesis is upheld, i.e. if the \( \kappa \) and \( s \) values for the different curves are comparable, the calculated \( \kappa \) and \( s \) values can be used to calculate the equation of the complete \( \ln t \) vs \( \ln v \) curve. Using equation (9), we get:

\[
\frac{dy}{dx} = \left[ \left( \frac{dy}{dx} \right)^2 - 1 \right]^{1/2}, \tag{10}
\]

where \( y = \ln t \) and \( x = \ln v \). This result can be used in equation (6) to calculate \( \Gamma(v)/y \). The reference volume \( v_0 \) is the basis volume for the calculation of the arc length \( s \).

The calculated \( \Gamma(v)/y \) function can then be used to calculate the similarity variable, \( \zeta \) and convert the \( F(v, t) \) experimental data into the self-similar distribution \( f(\zeta) \). This function is then used to calculate the daughter drop distribution via equation (7).

3.3. Determination of daughter drop distribution

Equation (7) for the determination of the daughter drop distribution is a Fredholm equation of the first kind and hence is an ill-posed problem, i.e. small changes in \( \xi'(\zeta) \) can cause large differences in the extracted \( \gamma g(x) \) function. Special techniques are needed to solve such ill-posed problems. These techniques make use of a priori information about the unknown function.

A widely used technique to solve ill-posed problems is Tikhonov regularization (Tikhonov and Arsenin, 1977; Wahba, 1977; Delves and Mohamed, 1985). This technique uses information about the smoothness of the unknown function. This becomes clear upon rewriting equation (7) in operator form (Ramkrishna and Amundson, 1985):

\[
\Phi = Kg. \tag{11}
\]

where \( \Phi \) is the vector representing the left-hand side of the equation, \( g \) is the vector representing the unknown function and \( K \) is the integral operator which acts on \( g \). In zero-order Tikhonov regularization, \( g \) is determined by the following:

\[
\min ||Kg - \Phi||^2 + \lambda_{reg} ||g||^2, \tag{12}
\]

where \( ||f|| \) is the norm and the subscripts 1 and 2 represent the possibility that different norms can be used. By using the norm of the unknown function in the minimization, Tikhonov regularization penalizes functions which fluctuate greatly and hence determines functions which are smooth.

However, in the case of equation (7), we have more information regarding \( g \) than that it is a smooth function. Since the daughter drop distribution, \( g(x) \), is a cumulative distribution function, it is a monotonic function of its argument. The set of bounded, monotonic functions is compact in the space \( L_2 \) (Tikhonov and Arsenin, 1977; Goncharskii and Jagola, 1969). Ivanov (1962) has shown that the method of quasisolutions can be used to find the solution of an ill-posed problem in this case. A quasisolution of equation (7) on a given compact set \( M \) of the space \( L_2 \) and for a given \( \Phi \) is the point \( g_0 \in M \) for which \( ||Kg - \Phi|| \) attains a minimum on \( M \). In this case, the compact set \( M \) is the set of monotonic functions. Using the method of quasisolutions, \( g \) is determined by:

\[
\min_{g \in M} ||Kg - \Phi||^2. \tag{13}
\]
The fact that \( g(x) \) is a cumulative distribution function allows us to determine the unknown constant \( \gamma \). Since at \( x = 1 \), \( g = 1 \), the solution of equation (7) for \( \gamma g(x) \) will allow us to determine \( \gamma \).

Solution of equation (13) proceeds by transforming it to a matrix minimization problem. The similarity coordinate \( \xi \) is discretized into a set of \( \{\xi_i\} \). Also, \( g(x) \) is expanded in terms of basis functions:

\[
y g(x) = \sum_{j=1}^{nb} a_j G_j(x).
\] (14)

where \( nb \) is the number of basis functions used in the expansion. Define the matrix \( X \) as:

\[
X_i = \int_0^1 x^i f'(\frac{\xi}{x}) G_j(x) dx. \tag{15}
\]

The term in the minimization [equation (13)] then becomes:

\[
a'X'Xa - 2a'X'\Phi + \Phi' \Phi \tag{16}
\]

where the superscript \( t \) represents the transpose of the matrix. Since the \( \Phi'\Phi \) term is independent of \( a \), the problem of determining the daughter drop distribution becomes the following constrained quadratic minimization problem:

\[
\min a'X'Xa - 2a'X'\Phi, \tag{17}
\]

to determine the unknown vector \( a \). The solution is constrained to be a monotonic increasing function.

The choice of discretization points and basis functions remain to be determined. These mainly depend on the nature of the similarity distribution. We need to choose the discretization points such that there are more points in the regions of \( \xi \) where there is maximum variation in the similarity distributions of different daughter drop distributions. By doing this, we give more weight to these regions in the inverse problem and hence, we will be able to discriminate successfully between different functions.

Similarity distributions for different \( y g(x) \) functions were determined, both analytically and by direct solution of the population balance equation, for further examination. Some of these distributions are shown plotted in Fig. 1. As shown in the figure, the distributions vary substantially in the size and position of the peak. The region under the peak contains the most information about the similarity distribution. In order to get more points in this region, we can choose our discretization intervals such that there are equal areas under the \( f'(\xi) \) curve or the \( \xi f'(\xi) \) curve in each interval. We have chosen discretization intervals having equal areas under the \( f'(\xi) \) curve.

The choice of basis functions is strongly dependent on the behavior of the similarity distribution. It can be shown that if \( \xi f'(\xi) \approx \xi^m \) for \( \xi \to 0 \), then \( g(x) \approx x^m \) for \( x \ll 1 \) (see Appendix). Expansion of \( \gamma g(x) \) in terms of orthogonal polynomials will converge very slowly to the desired function in cases where \( m < 0 \). Since in these cases \( g(x) \) is singular at \( x = 0 \). In order to overcome this difficulty, we choose basis functions which are problem-specific. We choose a space in which the inner product is given by the following equation:

\[
(u, v) = \int_0^1 x^{-m} u(x) v(x) dx. \tag{18}
\]

The orthogonal basis functions on such a space are modified Jacobi polynomials, \( x^m G_j(x) \).

Since the similarity distribution appears on both sides of equation (7), we need a straightforward method for representing the distribution. This will simplify the matrix equations for the determination of \( a \). Fitting the similarity distribution will also have the effect of limiting experimental error and will provide us a distribution which is continuous. As described by Wright and Ramkrishna (1992), the form to which the similarity distribution is fitted is a linear combination of gamma distributions. This form can exhibit all of the qualitative and quantitative behavior of the self-similar distribution. Hence, the form used for fitting the self-similar distribution is:

\[
\xi f'(\xi) = \sum_{k=1}^{\text{max}} A_k \xi^{k-1} \exp(-\beta_k \xi). \tag{19}
\]

The self-similar distribution data is fitted to the above expression using a weighted nonlinear regression fit. The weights used are estimates of the relative error at each point in the distribution. Techniques to estimate this error from the data are described by Silverman (1986).

Since an analytical form, equation (19), is used to represent the similarity distribution, the matrix \( X \) can be explicitly calculated. Let \( G_j \) denote the \( (j-1) \)th order Jacobi polynomial. Using equation (19) for the representation of \( \xi f'(\xi) \) and the known form of the Jacobi polynomials, we get:

\[
X(i, j) = \sqrt{2j} \sum_{m=1}^{\text{max}} A_m \sum_{k=1}^{\text{max}} (-1)^{m+1} \times \frac{(2j-m)!}{(m-1)!(j-m)!(j-m+1)!} \times \psi(a_m + m - j - a \beta_k \xi) \times \frac{\gamma_k(a_m + m - j - a \beta_k \xi)}{\beta_k^{m+1}}. \tag{20}
\]

where \( \psi \) is the complementary incomplete \( \gamma \)-function which is available in the SFUN library of IMSL and \( \mu \) is obtained from the behavior of the similarity distribution at small \( \xi \).
A constrained quadratic minimization program (LSSOL from the Stanford Office of Technology Licensing) is used to determine the unknown coefficients of expansion \( a \). The constraints used are physical constraints from the nature of the daughter drop distribution. The daughter drop distribution is positive everywhere and is monotonic increasing, as mentioned previously. We further surmise that break-up of a drop cannot produce drops in the neighborhood of zero volume and hence, drops nearly its own size, so that the derivative of the daughter drop distribution at \( x = 1 \) may be set equal to zero. This rather specific assumption may not apply to other break-up processes. Mathematically, the constraints used are:

\[
\begin{align*}
    g(x) &> 0 \\
    g'(x) &\geq 0, \\
    g'(1) &= 0.
\end{align*}
\]

(21)

The positivity and monotonicity constraints are implemented as a set of linear constraints on the coefficients of expansion such that positivity and monotonicity are maintained at every discretization point. The equality constraint is also implemented as a linear constraint on the coefficients at \( x = 1 \). In each case we have tested, 500 discretization points were used and three to seven basis functions were used to expand the unknown function.

4. EXAMPLE APPLICATIONS OF THE INVERSE PROBLEM

In this section, we present three examples of the inverse problem procedure. Two examples use known breakage rates and daughter drop distributions to compare the inverse problem results with the actual functions. In the third example, we apply the inverse problem technique to experimental data of transient drop size distributions in a stirred dispersion. In this case, the inverse problem results are evaluated by comparing the drop size distributions...
predicted by the extracted functions with the experimental data.

With the known breakage functions, we generated transient drop size distributions using a Monte-Carlo simulation procedure. The details of this simulation procedure are given in Ramkrishna et al. (1995). The initial drop size distribution was monodisperse. Since the data was generated by simulations, it inherently has some error. In both the cases presented, the breakage frequency function used was:

\[ \Gamma(v) = 1.2 \exp[0.12(\ln v + 3.5)] - 0.20(\ln v)^2 - 12.25, \]  

(22)

4.1. Case 1

The daughter drop distribution function used in this case was:

\[ g(x) = \frac{8}{3} x - \frac{5}{3} x^3. \]  

(23)

The simulated distributions were obtained for various times. The simulated data was then tested for the existence of similarity. The data was plotted as \( \ln t \) vs \( \ln v \) for 14 different values of \( F \). Figure 2 shows a plot of some of these curves. Each curve was fitted with a linear or quadratic form and the arc length and curvature of the curves was calculated from the fit. Figure 3 shows a plot of the arc length vs \( \ln v \) for the different curves. The figure clearly shows that the various arc length curves have collapsed into a single, smooth curve, thus upholding the similarity hypothesis. From this collapsed arc length curve, the breakage rate was calculated via equation (10). The result of this calculation is shown in Fig. 4 along with the actual function given by equation (22). From the figure it can be seen that the calculated breakage rate is very close to the actual function.
The extracted breakage rate function was then used to calculate the similarity distribution from the simulated data. Figure 5 shows the resulting distribution at six different times. It clearly shows that the data at different times have collapsed into a single curve. The data was fitted using equation (19). The results of the fit are $n_{\text{term}} = 2$, $A_1 = 0.46228$, $\beta_1 = 1.44229$, $A_2 = 0.20695$, $\alpha_2 = 1.43962$, $\beta_2 = 5.66412$. These parameters are used to calculate the discretization intervals by calculating equal areas under the $f'(\zeta)$ curve. Also, from these parameters and the discretization intervals, the $X$ matrix can be calculated. These are then used to determine the daughter drop distribution. This result is shown in Fig. 6. The different curves are the extracted daughter drop distributions for different numbers of basis functions.

In this case, it can be seen that the result with a small number of basis functions ($nb = 3, 4$) is very close to the actual function. As the number of basis functions increases, the result becomes more accurate. The effect of regularization on the daughter drop distribution extracted with 5 basis functions is shown in Fig. 7.
functions is increased, the result starts to deviate from the actual function. This result is expected. The similarity distribution is only known to some degree of certainty, i.e., there is some error in the distribution. With small numbers of basis functions, the inverse problem gives us a result which will predict the similarity distribution with the same order of magnitude of error. As \(n\) increases, the inverse problem can stay within the constraints imposed by the minimization technique and still determine a function which will predict the similarity distribution to a very high degree of accuracy.
Fig. 10. Comparison of breakage rate extracted from inverse problem with the actual function.

Since it is predicting the self-similar distribution to a much higher degree of accuracy than known from the data, the \( y_g(x) \) function determined deviates from the actual function.

This situation encountered with large \( nb \) is typical of those in which regularization is used to solve the ill-posed inverse problem. The inverse problem was tested with regularization for the case of \( nb = 5 \). We solved equation (12) with the constraints given by equation (21). The result is shown in Fig. 7. As we include regularization, the result is free from oscillations and converges towards the actual solution.

4.2. Case 2

The daughter drop distribution function used in this case was:

\[
g(x) = 2.5x^{0.3} - 1.5\sqrt{x}. \tag{24}
\]

As in the previous case, simulated distributions were obtained for various times. The simulated data was then tested for the existence of similarity. The data was plotted as \( \ln t \) vs \( \ln \sigma \) for 14 different values of \( F \). Figure 8 shows a plot of some of these curves. The arc length and curvature of the curves was calculated as in the previous case. Figure 9 shows a plot of the arc length vs \( \ln \sigma \) for the different curves. The figure clearly shows that the various arc length curves have collapsed into a single, smooth curve, thus upholding the similarity hypothesis. From this collapsed arc length curve, the breakage rate was calculated via equation (10). The result of this calculation is shown in Fig. 10 along with the actual function given by equation (22). From the figure it can be seen that the calculated breakage rate is very close to the actual function.

Figure 11 shows the resulting distribution at six different times. It clearly shows that the data at different times have collapsed into a single curve. The data was fitted using equation (19). The results of the fit are \( n_{term} = 2, A_i = 0.25543, \alpha_i = 1.25980, \)
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$\beta_1 = 1.44773, \ A_2 = 0.086692, \ \alpha_5 = 1.26026, \ \beta_2 = 9.68561$. These parameters are used to calculate the discretization intervals by calculating equal areas under the $f'(\zeta)$ curve. Also, from these parameters and the discretization intervals, the $X$ matrix can be calculated. These are then used to determine the daughter drop distribution. This result is shown in Fig. 12. The different curves are the extracted daughter drop distributions for different numbers of basis functions.

As in the previous case, the results with $nb = 3, 4$ are close to the actual function. The results with $nb = 5, 6, 7$ have again started deviating from the actual function. As discussed in Case 1, this is because as we increase the number of basis functions, the inverse problem can predict the data to a high degree of accuracy (which is much higher than the accuracy in the data) and still give results which are within the imposed constraints.

Since this behavior is similar to cases in which regularization is used, we tried to use regularization for the case with $nb = 5$. Figure 13 shows the unregularized result which deviates from the actual function along with the result obtained with a small regularization parameter. The small amount of

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Fig. 12. Daughter drop distribution calculated from the inverse problem for the second example.

Fig. 13. Effect of regularization on the daughter drop distribution extracted with 5 basis functions.

Fig. 14. Experimental drop size distributions at 500 rev min $^{-1}$ for a system of benzene-carbon tetrachloride in water.

Fig. 15. Breakage rate calculated from inverse problem using experimental data.
regularization allows the inverse problem to determine a function close to the actual function.

4.3. Case 3

Transient breakage experiments were carried out with a system of neutrally buoyant benzene and carbon tetrachloride in water. By using a very small amount of the organic phase, the drop size distribution evolves by pure breakage for some time. The data used in this example, shown in Fig. 14, were obtained by stirring the system at 500 rev min⁻¹.

From the plots of In t vs In v for 14 different values of F, it was found that the curves all collapsed into one, upholding the similarity hypothesis. From the collapsed curve, the breakage rate was calculated. The resulting breakage rate is shown in Fig. 15. The resulting similarity distribution is shown in Fig. 16. As shown in the figure, the data at various times have collapsed into a single curve. This similarity distribution is then used to calculate the daughter drop distribution. The results for different numbers of basis functions are shown in Fig. 17.

In order to test the results obtained from the inverse problem, we used the extracted breakage rate and daughter drop distribution functions to predict the transient size distributions. The initial condition for the solution of the population balance equation was taken to be the distribution at the time at which similarity is first observed. For the case of the daughter drop distribution determined with 3 basis functions, the predicted distributions in terms of the volume fraction density are shown in Fig. 18 along with the experimental data. From the figure, it can be seen that the transient distributions are predicted very well by the extracted functions.
5. EXPERIMENTAL UNCERTAINTY AND CHOICE OF NUMBER OF BASIS FUNCTIONS

From the preceding examples, it can be seen that the choice of number of basis functions plays a crucial role in determining the inverse problem result. The examples also give us guidelines on how to make this choice. We should choose the smallest number of basis functions such that the result is consistent with the uncertainty in the similarity distribution. By doing so, we will not be giving undue importance to error in the data.

Since equation (19) is used to fit the similarity distribution, we can calculate the sum square error of the fit to the experimental data. This gives us an estimate of the error in the data. Since the fitted distribution is used to calculate the daughter drop distribution, we should expect the extracted function to predict the fitted distribution to the same level of accuracy that the fit predicts the data. Using the extracted function, $g(x)$ and equation (15), we can calculate the $c[f(x)]$ value predicted by the $g(x)$ function at each of the experimental data points and hence, calculate the sum square error in these predictions to the fitted distribution. We then choose the smallest number of basis functions such that this error is not more than the estimated error in the data.

These guidelines are used on the three example cases described in the previous section. For the first
case, Table 1 gives the sum square error for the fit of the data and the error for each of the extracted functions. From the data in the table, it can be seen that the result with 3 basis functions predicts the data within the experimental error. Figure 6 confirms that the result with 3 basis functions is close to the actual function.

Table 2 gives the sum square error for the fit of the data and for each of the extracted functions for the second case. In this case, it can be seen that the smallest number of basis functions required to predict the data within experimental error is 3. Figure 12 confirms that the result with 3 basis functions predicts the actual function quite well.

Table 3 gives the sum square error for the fit of the data and the error for each of the extracted functions for the third example. In this case, we see that the result with 3 basis functions predicts the data within experimental error. Figure 18 confirms that the result with 3 basis functions predicts the experimental data well.

6. SUMMARY AND CONCLUSIONS

A mathematical and computational technique for the determination of drop breakage rate and daughter drop distribution from transient size distributions has been given. The technique makes use of a similarity transformation of the population balance equation.

The choice of similarity variable is described and a test for the existence of similarity is outlined. The technique makes use of a similarity transformation of the population balance equation.

The choice of similarity variable is described and a test for the existence of similarity is outlined. The inverse problem technique uses the method of quasi-solutions to determine the daughter drop distributions. A technique for the choice of number of basis functions in this procedure is described. Using these techniques, good estimates of the breakage rate and daughter drop distribution have been extracted from computer simulated data.

The inverse problem procedure has also been successful in the determination of droplet breakage rates and daughter drop distributions in stirred liquid–liquid dispersions (Sathyagal et al., 1995) from experimental data.

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REFERENCES


APPENDIX

Asymptotic Behavior of the Daughter Drop Distribution

The equation for the similarity distribution, equation (7) can be rewritten as follows:

\[ \xi f' (\xi) = \int_0^\infty g\left( \frac{x}{\xi} \right) \xi f'(\xi') \, d\xi'. \]  

(25)

Let us denote \( \xi f'(\xi) \) by \( \phi(\xi) \).

In all our experimental data, we have observed that the behavior of the similarity distribution at small \( \xi \) can be described by:

\[ \phi(\xi) = c \xi^\alpha. \]  

(26)

where \( c \) is a constant. We are interested in determining the behavior of \( g(x) \) for \( x \ll 1 \), given this behavior of \( \phi(\xi) \) at small \( \xi \).
Let $\zeta$ belong to the region where equation (26) is applicable, and define $\zeta_{m}$ such that:

$$\zeta_{m}(1-h)<\zeta<\zeta_{m}, \quad h \neq 1.$$  

(27)

Equation (25) can be written as:

$$\phi(\zeta) - \gamma \int_{\zeta_{m}}^{\infty} g\left(\frac{x}{\zeta}\right) \phi(x) dx - \frac{g'(1)}{2} \int_{\zeta_{m}}^{\infty} \left(1 - \frac{x}{\zeta}\right) \phi(x) dx - \gamma \int_{\zeta_{m}}^{\infty} g\left(\frac{x}{\zeta}\right) \phi(x) dx.$$  

(28)

For the first term on the right-hand side of equation (28), we can expand the $g(x)$ function in a Taylor series around $x = 1$ as:

$$g(x) = 1 - g'(1)(1 - x) + \frac{g''(1)}{2} (1 - x)^2 + \cdots$$

Since we are dealing with small values of $\zeta$ and $\zeta_{m}$, equation (26) can be substituted in the left-hand side and the first term on the right-hand side of equation (28). This equation can then be written as:

$$c\zeta^{m} = -c\gamma \int_{\zeta_{m}}^{\infty} \left(1 - g'(1)(1 - x) + \frac{g''(1)}{2} (1 - x)^2 + \cdots\right) \zeta^{m-1} dx$$

$$+ \frac{g'(1)}{2} \int_{\zeta_{m}}^{\infty} \left(1 - \frac{x}{\zeta}\right) \zeta^{m-1} dx$$

$$+ \cdots + \gamma \int_{\zeta_{m}}^{\infty} g\left(\frac{x}{\zeta}\right) \phi(x) dx.$$  

(29)

This equation can be differentiated to obtain:

$$c\mu \zeta^{m-1} = -c\gamma \int_{\zeta_{m}}^{\infty} \left(1 - g'(1)(1 + \cdots) + g''(1) \left(\frac{h}{2} + \cdots\right) + \cdots\right) \zeta^{m-1} dx$$

$$+ \gamma \int_{\zeta_{m}}^{\infty} g\left(\frac{x}{\zeta}\right) \phi(x) dx.$$  

(30)

Performing the indicated integrations, we get:

$$c\mu \zeta^{m-1} = -c\gamma \left[1 - g'(1)(1 + \cdots) + g''(1) \left(\frac{h}{2} + \cdots\right) + \cdots\right]$$

$$+ \gamma \int_{\zeta_{m}}^{\infty} g\left(\frac{x}{\zeta}\right) \phi(x) dx.$$  

(31)

This can be written as:

$$c\zeta^{m-1} \left[\mu + \gamma \zeta [1 + O(h)]\right] = \gamma \int_{\zeta_{m}}^{\infty} g\left(\frac{x}{\zeta}\right) \phi(x) dx.$$  

(32)

Using $\zeta = \zeta_{m}[1 + O(h)]$, we can rewrite the above equation as:

$$c\zeta^{m} + O(h) = \gamma \int_{\zeta_{m}}^{\infty} g\left(\frac{x}{\zeta}\right) \phi(x) dx.$$  

(33)

Similarly, from equation (29), we get:

$$c\zeta^{m} + O(h) = \gamma \int_{\zeta_{m}}^{\infty} g\left(\frac{x}{\zeta}\right) \phi(x) dx.$$  

(34)

Substituting for $c\zeta^{m}$ from equation (34) into equation (33), we get:

$$\gamma \int_{\zeta_{m}}^{\infty} g\left(\frac{x}{\zeta}\right) \phi(x) dx + O(h)$$

$$= \gamma \frac{\zeta}{\mu + \gamma m} \int_{\zeta_{m}}^{\infty} \frac{g\left(\frac{x}{\zeta}\right)}{\zeta} \phi(x) dx + O(h).$$  

(35)

Hence, the leading behavior of $g(x)$ can be determined from:

$$\gamma \int_{\zeta_{m}}^{\infty} \left[1 - \frac{x}{\zeta} \right] \phi(x) dx = \frac{\zeta}{\mu + \gamma m} \int_{\zeta_{m}}^{\infty} \left[1 - \frac{x}{\zeta} \right] \phi(x) dx.$$  

(36)

Since equation (36) has to hold for every $\zeta$ for which equation (26) is valid, the integrand must be identically zero. Hence:

$$\gamma \int_{\zeta_{m}}^{\infty} \left[1 - \frac{x}{\zeta} \right] \phi(x) dx = \frac{\zeta}{\mu + \gamma m} \int_{\zeta_{m}}^{\infty} \left[1 - \frac{x}{\zeta} \right] \phi(x) dx = 0.$$  

(37)

This equation can be solved to obtain:

$$g(x) = c_{1}, x^{\mu} \gamma^{m}.$$  

(38)

Since $\zeta_{m}$ is usually small, by choosing $\zeta_{m} \approx \mu / \gamma$, we find that the leading behavior of $g(x)$ is given by:

$$g(x) = c_{1}, x^{\mu}.$$  

(39)

Experimentally, the $\mu$-values we have found from the behavior of the similarity distribution at low $\zeta$ have been in the range 0.2 to 0.8 and the $\gamma$-values have been of the order of 1. Since the small $\zeta$ behavior of the similarity distribution given by equation (26) is in the region $\zeta \ll 1$, the assumption that $\zeta_{m} \approx \mu / \gamma$ is valid and equation (39) gives us the leading behavior of the daughter drop distribution.

Analytical expressions for the similarity distribution can be determined from solution of equation (25) for two particular forms of the $g(x)$ function. In both these cases, we find that the relation determined by the above analysis between the behavior of the similarity distribution at low $\zeta$ and the leading behavior of $g(x)$ is upheld.

If $g(x) = x^{\nu}$, equation (25) can be written as:

$$\phi(\zeta) = \frac{\mu}{\gamma^{m}} \zeta^{\nu} \exp(-\gamma \zeta).$$  

(40)

This equation can be solved to get:

$$\phi(\zeta) = \frac{\mu}{\Gamma(\mu)} \zeta^{\nu} \exp(-\gamma \zeta).$$  

(41)

where $\Gamma(\cdot)$ refers to the gamma function. Hence, in this case, for small $\zeta$ we find that $\phi(\zeta) \sim \zeta^{\nu}$.

Another analytical solution can be obtained for $g(x) = \alpha x^{\beta} + (1 - a) x^{\beta}$. Ziff (1991) has solved an equation similar to equation (25) for this form of $g(x)$. Following his solution, we obtain in this case:

$$\phi(\zeta) = \frac{\Gamma(1 - a) \beta + a \beta}{\Gamma(\beta + 1) \Gamma(\alpha)} \zeta^{a \beta} \exp(-\gamma \zeta).$$  

(42)