On Evolution of Drop-Size Distributions in Turbulent Pipe Flow Revisited

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This paper corrects an error in the work of Nere and Ramkrishna [Ind. Eng. Chem. Res. 2005, 44, 1187] on solving population balance equations for the evolution of drop-size distributions in fully developed turbulent flow of a liquid–liquid dispersion in a circular pipe. We rely on the previously noted Nere and Ramkrishna’s work for the formulation of the problem. An alternative linear operator formulation is developed here to solve the population balance equation, compared with detailed numerical solution obtained by computational fluid dynamics (CFD) and is determined to be considerably more efficient.

1. Introduction

A recent paper by Nere and Ramkrishna1 presented a solution to the population balance equation that governs the spatial evolution of drop-size distributions for a liquid–liquid dispersion in fully developed turbulent flow, by breakage of the droplets caused by hydrodynamic forces. The droplets were allowed to diffuse only radially, because axial diffusion was judged to be unimportant, compared with axial convective displacement. Droplet breakage frequency, which is presumed to be dependent on the local energy dissipation, is then a function of the radial coordinate; however, the daughter drop-size distribution becomes spatially independent through a scaling behavior that was originally described by Narsimhan et al.2 The solution used two basic features: one the transformation of spatial coordinates into a temporal coordinate, and the second the identification of a linear, self-adjoint (diffusion-like) differential operator whose eigenfunctions were used to expand the solution. This solution is inadmissible in the presence of the diffusion of particles. However, we show here that an elegant linear operator method can indeed be used to solve the problem, retaining the spatial dependence of the breakage rate, as in the paper of Nere and Ramkrishna.3 Furthermore, in computations, the Reynolds number of the flow and the breakage rate are increased sufficiently to introduce significant diffusion and breakage, in contrast with the work of Nere and Ramkrishna,1 which prevented numerical diagnosis of the error in question in their paper.

2. Operator Solution to Population Balance Equation

No background discussion of the problem setting is included here, because it remains the same as that described earlier. However, we cast the problem in somewhat different dimensionless form, which is more consistent with past convention in the chemical engineering literature. Defining the Peclet number \( Pe \equiv Ru/V \), where \( R \) is the pipe radius and \( u \) and \( V \) are the average axial velocity and diffusion coefficient, respectively) and \( \beta \equiv R^2 V / D \) (which compares the diffusion time to the breakage time), the conservation equation in fully developed turbulent flow with a (dimensionless) velocity profile \( u(r) \) may be written directly for the discretized population balance equation as follows:

\[
\frac{\partial N_i(r,z)}{\partial z} - \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial N_i(r,z)}{\partial r} \right) - \sum_{k} \gamma_{ik} \Gamma_N(r,z) N_i(r,z) = \beta \Gamma_N(r,z) AN(r,z) \quad (for \ i = 1, 2, ..., n)
\]

where the discretization coefficient \( n_{ik} \) that was used by Nere and Ramkrishna1 has been replaced by \( \gamma_{ik} \). The foregoing equation may be rewritten in more-compact vector form as

\[
Pe_v \frac{\partial \mathbf{N}(r,z)}{\partial z} - \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial \mathbf{N}(r,z)}{\partial r} \right) = \beta \Gamma_N(r,z) \mathbf{AN}(r,z)
\]

Following Nere and Ramkrishna,1 the boundary conditions may be written for \( \mathbf{N} \) as

\[
\frac{\partial \mathbf{N}}{\partial r} = 0 \quad (for \ r = 0, 1)
\]

The linear operator to solve the foregoing problem is obtained through the following definitions. First, we define the \( n \)-dimensional Hilbert space \( \mathcal{H} \) in which the inner product is the usual inner product between any two vectors \( x \equiv (x_1, x_2, ..., x_n) \) and \( y \equiv (y_1, y_2, ..., y_n) \), defined by

\[
\langle x, y \rangle = \sum_{i=1}^{n} x_i y_i \quad (for \ x, y \in \mathcal{H})
\]

Next, we define the Hilbert space \( \mathcal{H} \equiv L^2([0,1], ru(r) \) of functions in which the inner product between any two functions \( u \) and \( v \) is defined by

\[
\langle u, v \rangle = Pe \int_{0}^{1} ru(r)u(r)v(r) \, dr \quad (for \ u, v \in \mathcal{H})
\]

Next, we define the tensor product Hilbert space \( \mathcal{H} \equiv \mathcal{H} \otimes \mathcal{H} \) (See Ramkrishna and Amundson3 for a definition of the tensor product.) The vector \( \mathbf{N}(z) \equiv (\mathbf{N}(r,z)) \) is an element of \( \mathcal{H} \) because it represents the \( n \) number densities of the discrete particle sizes, each representing a vector in the Hilbert space, \( \mathcal{H} \) for a fixed \( z \). For an operator formulation of the problem, we define the following operators. Let \( L_r = \{L_r(DL_r)\} \), where
\[
L_r = \left[ -\frac{Pe^{-1}}{ru(r)} \frac{d}{dr} \left( \frac{d}{dr} r \varphi(r) \right) \right]
\]

\[
D(L_r) = \{ u \in \mathcal{H}; \ L u \in \mathcal{H}; \ u^0 = u'(0) = 0 \}
\]

The boundary conditions are clearly absorbed by the domain of the operator \( L_r \), which is a subspace of \( \mathcal{H} \). Furthermore, we define the operator, \( B_r = \beta \varphi(r) Peu(r) \), where \( I \) is the identity operator on \( \mathcal{H} \). In what follows, it is essential to recognize the identity matrix operator \( I \) and the matrix operator \( A \), which were defined earlier from the discretized breakup process as both acting on the Hilbert space \( \mathcal{H} \). Finally, to represent the boundary value problem of interest, we define the operator \( L \) on the tensor product Hilbert space \( \mathcal{H} \) by

\[
L \equiv L_r \otimes I + B_r \otimes A
\]

The entire boundary value problem (eqs 2 and 3) then can be written as

\[
\frac{\partial N(z)}{\partial z} = -LN(z)
\]

the solution of which may be written as

\[
N(z) = \exp(-zL)N(0)
\]

The exponential operator \( \exp(-zL) \) from a suitable spectral representation of \( L \) now remains to be interpreted. Because the matrix \( A \) is upper triangular, it has a representation that is given by

\[
A = \sum_{k=1}^{n} \lambda_k S_k
\]

where \( \{\lambda_k\} \) are the eigenvalues of the matrix \( A \), which are readily determined to be its diagonal elements. The matrices \( \{S_k\} \) are the projection matrices as appearing in Sylvester’s expansion (see Amundson) satisfying the property, \( \sum_{k=1}^{n} S_k = I \). The matrix \( S_k \) may be elegantly represented as

\[
S_k = \frac{v_k w_k^T}{w_k^T v_k}
\]

where \( v_k \) and \( w_k^T \) are, respectively, column and row eigenvectors of \( A \) corresponding to eigenvalue \( \lambda_k \). Thus, the operator \( L \) can now be written as

\[
L = \sum_{k=1}^{n} (L_r + \lambda_k B_r) \otimes S_k = \sum_{k=1}^{n} L^{(k)}_r \otimes S_k
\]

It is readily observed that the operator \( L^{(k)}_r \equiv (L_r + \lambda_k B_r) \) is self-adjoint and its spectral representation may be written as

\[
(L_r + \lambda_k B_r) = \sum_{i=1}^{\infty} \mu_i^{(k)} P_i^{(k)}
\]

where the \( \{\mu_i^{(k)}\} \) are the eigenvalues of \( L^{(k)}_r \) and \( \{P_i^{(k)}\} \) are the projection operators, which must be determined computationally and is discussed subsequently. The spectral expansion described by eq 7 implies that the exponential operator \( \exp(-zL) \) has the expansion

\[
\exp(-zL) = \sum_{k=1}^{\infty} e^{-\mu_i^{(k)} z} (P_i^{(k)} \otimes S_k)
\]

Thus, the operator solution to the problem is readily written as

\[
N(z) = \sum_{k=1}^{\infty} e^{-\mu_i^{(k)} z} (P_i^{(k)} \otimes S_k) N(0)
\]

Because the eigenvalue problem cannot be solved analytically, the solution was obtained numerically using the Arnoldi algorithm (Saad), with the aid of Matlab. Sample eigenvalues that correspond to \( \lambda = 1 \) are 0.0129, 0.0907, 0.2340, 0.4408, 0.7121, 1.0467, 1.4468, 1.9132, 2.4466, 3.0516, and 3.7264, whereas Figure 1 shows eigenfrequencies that correspond to the first five eigenvalues. We made certain that the eigenfrequencies flatten out very close to the wall, although this feature may not be prominent in the figure, leading to zero gradient. The reader may note that the solution could have been done for the case of size-dependent diffusion with more computation time.

The direct solution approach toward solving the population balance (eq 2) is to integrate the equation in space using control volume approach, as elaborated in Nere and Ramkrishna. The grid size used in the present computation was 100 \( \times \) 5000. The grid size used in the present computation was 100 \( \times \) 5000.

2.1. Cup-Mixed Average Drop Size at the Pipe Outlet. The calculation of the (dimensionless) cup-mixed average drop size at any axial location \( z \) of the pipe, denoted as \( \langle X(z) \rangle \), is obtained in an elegant way. Recognizing the definition of the cup-mixed average particle size, one obtains

\[
\langle X(z) \rangle = \frac{\sum_{k=1}^{n} \int_0^1 ru(r) N_k(r,z) \, dr \cdot \sum_{k=1}^{n} v_k (1 N_k(z))}{\sum_{k=1}^{n} \int_0^1 ru(r) N_k(r,z) \, dr \cdot \sum_{k=1}^{n} (1 N_k(z))} = \frac{\langle 1 \sum_{k=1}^{n} v_k N_k(z) \rangle}{\langle 1 \sum_{k=1}^{n} N_k(z) \rangle} = \langle 1 \langle X^2 \rangle N(z) \rangle
\]

where \( 1 \) represents the function that is unity in the interval from \( r = 0 \) to \( r = 1 \) and the boldface notation on \( N_k(z) \) applies to the number density of the \( k \)th bin size, which is viewed as an element of \( \mathcal{H} \); furthermore, it is the \( k \)th component of the vector \( N(z) \).

3. Comparison of Solutions

In this section, the focus is on comparison of the solution of the population balance (eq 2) by the linear operator formalism with that produced by the control volume approach. Table 1 displays the pipe diameter and the operating conditions used in the calculations. For the purpose of comparison, simulations were performed using monodispersed feed drop-size distribution at the inlet of the pipe (see Figure 2a). Figure 2b depicts the polydispersed feed drop-size distribution (normally distributed around the mean dimensionless drop size of 0.5 with a standard deviation of unity) that was used as another feed condition used to study the drop size evolution. The dimensionless number density plotted in figures refers to the number of the drops...
belonging to the corresponding size bin made dimensionless by the total number of initial drops per unit volume of space.

The droplet volume was discretized using the geometric grid with the number of bins \( n \) equal to 30 and consecutive volume ratio \( x_{i+1}/x_i \) of 1.3. The "exact" solution was constructed using the first 91 eigenvalues and eigenvectors. The word "exact" associated with the linear operator solution is shown in quotes to indicate its limitation in regard to being numerically exact!

Comparisons of the drop-size distribution from the solution given by eq 9 with that obtained from the direct solution...
approach, at two radial locations ($r = 0.8$ and $0.93$) and two axial locations ($z = 50$ and $100$) are shown in Figure 3a–d, which show close agreement between the two. The locations for comparison have been chosen to be significantly different with respect to the local flow, to emphasize the universality of agreement between the two solutions.

All the computations were performed on Pentium 4 computer with 1.3 GHz central processing unit (CPU). Note that the processor used is different than that used in the earlier paper. The comparison of the CPU times is given in Table 2. The time shown for operator solution corresponds to the calculation of number densities across a cross section of a pipe that includes the major time required to compute eigenvalues and eigenfunctions. The calculation at any other cross section (i.e., $z$) requires simple summation operation, requiring negligibly small time, as eigenvalues and eigenfunctions are already determined. It is clear that the operator solution obtains the drop size distribution with considerably shorter computation time than the direct solution approach. Furthermore, the operator method does not require additional computation to handle different feed conditions, which is an advantage that is unavailable to the direct solution approach.

4. Evolution of Drop Size Distribution in a Fully Developed Turbulent Flow Field

Figures 4–6 display the evolution of the drop-size distribution in a fully developed turbulent flow. Figures 4a and 4b show the size distributions at different radial locations at two different axial locations ($z = 50$ and $100$, respectively) for the monodispersed feed. It may be noted that the peak number density corresponds to almost the same drop volume at different radial locations. This interesting feature is a consequence of the redistribution of the drops, because of diffusion. Furthermore, one can see a drastic change in number densities, corresponding to the lower drop sizes for a small change in radial position close to the wall. This is because the energy dissipation rate changes with a steep gradient near the wall, which leads to the wider variation breakage rates. In addition, the time spent by the drops at a given axial location in the breakage environment is greater near the wall, because of lower velocities, which further contributes to an overall increase in breakage.

Figures 5a and 5b illustrate size distributions that evolve along the axial coordinate for a given radial location near the pipe center for the monodispersed and polydispersed feed conditions, respectively. Figures 6a and 6b demonstrate the same behavior near the wall. The profiles indicate the effect of diffusion of smaller drops from the periphery toward the center. The almost-

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*Table 2. Details of the CPU Time for the Simulations*

<table>
<thead>
<tr>
<th>task</th>
<th>CPU time (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>direct solution with control volume approach</td>
<td>119</td>
</tr>
<tr>
<td>operator solution</td>
<td>16 + $T^a$</td>
</tr>
</tbody>
</table>

$^a$ Here, $T$ is the computational time required for the construction of the number density profiles at any cross section of the pipe and is negligibly small.
constant number density along the axial coordinate at $x = 0.3$ for the radial location $r = 0.2$, which is depicted by Figure 5b, is a consequence of this diffusion effect.

It is of interest to note that the change in the drop-size distribution that is introduced because of the large change in axial position is relatively small, as compared to that introduced due to the change in radial position at a given axial location (Figures 4–6). This is of course because of the profiles of velocity and the energy dissipation rate, which enhance residence as well as breakage rate in breakage zones.

5. Conclusions

This paper has developed an “exact” solution for the population balance equation for the case of pure breakage in a fully developed pipe flow using linear operator theory. The breakage frequency is allowed to vary spatially but satisfies the similar breakage assumption of Narsimhan et al. and Sathyagal et al.

Acknowledgment

The authors are grateful to M. Kostoglou for pointing out the error in the paper by Nere and Ramkrishna.

Nomenclature

**A** = upper triangular matrix of elements given by breakage rates

**I** = identity matrix operator

**L** = identity matrix operator on the Hilbert space, $H$

$i, k =$ droplet size bin indices

$n =$ number of size bins used for the discretization of the particle state vector (i.e., drop volume)

$\gamma_{i,k} =$ redistribution coefficients in the discretization method of Kumar and Ramkrishna

$N(r,z) =$ discrete number density corresponding to the $i$th size bin in space (m$^{-3}$)

$N(r,z) =$ $n$-dimensional population density vector;

$N(z) =$ population density vector as an element of the Hilbert spaces, $H$

$P^{(k)} =$ projection operator (see eq 7)

$Pe =$ Peclet number; $Pe = \frac{R_i u_f}{\nu}$

$r =$ dimensional radial coordinate; $r = r/R$

$R =$ radius of the pipe (m)

$S_i =$ projection matrices as appearing in Sylvester’s expansion

$u_i =$ dimensionless axial velocity

$u_i =$ average mean axial velocity (m/s)

$v_i =$ right eigenvector

$x =$ dimensionless drop volume

$x =$ size vector consisting of drop sizes as elements

$X =$ dimensionless cup-mixed average drop size

$w_k =$ left eigenvector

$z =$ dimensionless axial distance; $z = z/R$

Greek Letters

$\beta =$ ratio of average diffusion time scale to breakage time scale;

$\beta \equiv \frac{R_i}{\nu} \left( \frac{\rho}{\rho_i} \right)^{1/2}$

$\tilde{\Gamma}_X =$ size-dependent breakage rate evaluated using average drop volume

$\bar{\Gamma}_X =$ space-dependent breakage rate evaluated using the average turbulent energy dissipation rate in the case under consideration

$\bar{\Gamma}_X =$ dimensionless size dependent breakage rate; $\bar{\Gamma}_X \equiv \frac{\gamma}{\bar{\gamma}}$

$\bar{\Gamma}_R =$ dimensionless space dependent breakage rate; $\bar{\Gamma}_R \equiv \bar{\gamma} \vec{r}$

$\vec{\lambda}_i =$ eigenvalue of a matrix $A$

$\vec{\mu}_i =$ eigenvalue of the operator $L_i$

Scripted Font Symbols

$\mathcal{B} =$ dimensionless droplet diffusivity

$\mathcal{D} =$ average diffusion coefficient over the pipe cross section (m$^2$/s)

$H =$ Hilbert space

$H_i =$ Hilbert space defined by $H_i \equiv L^2([0,1];ru_i(r))$

$\mathcal{R} =$ $n$-dimensional real vector space

Literature Cited


Received for review January 26, 2006

Revised manuscript received August 24, 2006

Accepted August 30, 2006

IE0601145