

The Bivariate Spatially distributed Population Balance Equation: An Accurate Reduction Technique

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Abstract

In this work, the advantages of the generalized fixed pivot technique as extended to mass transfer and the quadrature method of moments are utilized to reduce the bivariate spatially distributed population balance equation describing the coupled hydrodynamics and mass transfer in liquid-liquid extraction columns. The proposed reduction technique is found to reduce the discrete system of partial equations from $2M_x+1$ to M_x+2 , where M_x is the number of pivots or classes. The spatial variable is discretized in a conservative form using a couple of recently published central difference schemes. The numerical predictions of the detailed and reduced models are found almost identical accompanied by a substantial reduction of the CPU time as a characteristic of the reduced model.

1. Introduction

Liquid-liquid extraction columns (LLEC) are one of the major multiphase processes that call for population balance framework as a modeling tool due to their dispersed nature. This framework would help in the optimal design of such equipment that has not yet been fulfilled and still dependent on the time consuming and expensive scale up methods from laboratory scale pilot plants. In LLECs, the basic interactions such as droplet breakage and coalescence coupled to the interphase mass transfer result in a spatially distributed population of droplets with respect to the droplet size as well as concentration. In contrast to the previous spatially distributed population balance equation (SDPBE) describing the performance of LLECs (Mohanty, 2000), the present modeling approach allows the dynamic interaction of the mass transfer and fluid hydrodynamics by leaving it open to introduce a suitable model for predicting the interfacial tension, which changes as a function of solute transfer and markedly affects the breakage and coalescence rates (Attarakih et al., 2004).

In the present work, the state of any droplet is represented by a bivariate (joint) density function $n_{d,c_y}(d, c_y; t, z)$, where $n_{d,c_y}(d, c_y; t, z) \partial d \partial c_y$ represents the number of droplets having size and concentration in the ranges $[d, d+\partial d] \times [c_y, c_y+\partial c_y]$ per unit volume of

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the contactor. This allows the discontinuous macroscopic (breakage and coalescence) and the continuous microscopic (interphase mass transfer) events to be coupled in a single SDPBE along with the transport equations describing the hydrodynamics and mass transfer of the continuous phase. These equations represent a system of mixed integro-partial and algebraic equations for which no analytical solution exists except for strongly simplified forms, and hence a numerical solution is required in general. A detailed numerical algorithm based on the quadrature method of moments (QMOM) and the generalized fixed-pivot technique as extended to mass transfer (GFPMT) is presented by Attarakih et al. (2004a). The GFPMT could be viewed as a hybrid technique that couples the QMOM and the generalized fixed-pivot, which results in a two-population balance equation in terms of the droplet number and solute concentrations. Upon discretization using the GFPMT, this system of equations comprise a large system of conservation laws that is hyperbolically dominant. To reduce the dimensionality of the problem at hand a reduction technique is proposed in this work that is still based on the QMOM and the GFPMT. This reduction technique retains the whole information furnished by the number concentration function and averages out the solute concentration coordinate without any simplifying assumptions regarding the form of the bivariate density function. The numerical results for the simulation of a pilot plant RDC column shows that there is a significant reduction in the CPU time when compared to the detailed model.

2. The mathematical model

The general SDPBE for describing the coupled hydrodynamics and mass transfer in LLECs in a one spatial domain could be written as:

$$\begin{aligned} \frac{\partial n_{d,c_y}(\psi)}{\partial t} + \frac{\partial [u_y n_{d,c_y}(\psi)]}{\partial z} + \sum_{i=1}^2 \frac{\partial [\dot{\zeta}_i n_{d,c_y}(\psi)]}{\partial \zeta_i} = \\ \frac{\partial}{\partial z} \left[D_y \frac{\partial n_{d,c_y}(\psi)}{\partial z} \right] + \frac{Q_y^{in}}{A_c} n_y^{in}(d, c_y; t) \delta(z - z_y) + R\{\psi\} \end{aligned} \quad (1)$$

In this equation the components of the vector $\psi = [d \ c_y \ z \ t]$ are those for the droplet internal coordinates (diameter and solute concentration), the external coordinate (column height), z , and the time, t , where the velocity vector along the internal coordinates is given by $\dot{\zeta} = [\dot{d} \ \dot{c}_y]$. The source term $R \cdot \partial \zeta$ represents the net number of droplets produced by breakage and coalescence per unit volume and unit time in the coordinates range $[\zeta, \zeta + \partial \zeta]$. The left hand side is the continuity operator in both the external and internal coordinates, while the first part on the right hand side is the droplets axial dispersion characterized by the dispersion coefficient, D_y , which might be dependent on the energy dissipation and the droplet rising velocity. The second term on the right hand side is the rate at which the droplets entering the LLEC with volumetric flow rate, Q_y^{in} , that is perpendicular to the column cross-sectional area, A_c , at a location z_y with an inlet number density, n_y^{in} , and is treated as a point source in space. The dispersed phase velocity, u_y , relative to the walls of the column is determined in terms of the relative (slip) velocity with respect to the continuous phase (Attarakih et al., 2004a). The solute concentration in the continuous phase, c_x , is predicted using a component solute balance on the continuous phase.

3. Model Reduction

The reduction technique proceeds in two steps: **First** the quadrature method of moments (QMOM) is applied to integrate out the solute concentration after multiplying Eq.(1) by $c_y^m v(d)$ (with $m = 0$ and 1) and integrating it from 0 to $c_{y,max}$, which results in two

coupled marginal densities: $n_d(d, z, t)$ and $q(d, z, t) = \int_0^{c_{y,max}} c_y v(d) n_{d,c_y}(\psi) \partial c_y :$

$$\frac{\partial n_d(d; z, t)}{\partial t} + \frac{\partial}{\partial z} \left[u_y n_d(d; z, t) - D_y \frac{\partial n_d(d; z, t)}{\partial z} \right] = \frac{Q_y^{in}}{A_c} \frac{n^{in}}{v_{in}}(t) \delta(z - z_y) + \pi_n(n_d, \bar{c}_y) \quad (2)$$

$$\begin{aligned} \frac{\partial q(d; z, t)}{\partial t} + \frac{\partial}{\partial z} \left[u_y q(d; z, t) - D_y \frac{\partial q(d; z, t)}{\partial z} \right] = \\ \frac{Q_y^{in}}{A_c} \frac{c_y^{in} n^{in}(t)}{v_{in}} \delta(z - z_y) + \int_0^{c_{y,max}} \dot{c}_y n_{d,c_y}(d, c_y; z, t) \partial c_y + \pi_q(n_d, q, \bar{c}_y) \end{aligned} \quad (3)$$

The expressions of the source terms π_n and π_q are a little bit complex and are presented in Attarakih et al. (2004a). The application of the generalized fixed pivot technique to the system of equations (2) and (3) results in the following set of discrete PDEs:

$$\frac{\partial \varphi_i(z, t)}{\partial t} + \frac{\partial}{\partial z} \left[u_{y,i} \varphi_i(z, t) - D_y \frac{\partial \varphi_i(z, t)}{\partial z} \right] = \frac{Q_y^{in}}{A_c} \varphi_i^{in}(t) \delta(z - z_y) + \pi_{n,i} \quad (4)$$

$$\begin{aligned} \frac{\partial \Theta_i(z, t)}{\partial t} + \frac{\partial}{\partial z} \left[u_{y,i} \Theta_i(d; z, t) - D_y \frac{\partial \Theta_i(d; z, t)}{\partial z} \right] = \\ \frac{Q_y^{in}}{A_c} c_y^{in} \varphi_i^{in}(t) \delta(z - z_y) + \frac{6K_{oy,i}}{d_i} (\varphi_i c_y^*(c_x) - \Theta_i) + \pi_{q,i} \end{aligned} \quad (5)$$

Where $\varphi_i(z, t)$ and $\Theta_i(z, t)$ are the total volume and solute concentrations in the i th subdomain (interval). The mean solute concentration in the dispersed phase is obtained by combining the last two quantities as follows:

$$\bar{c}_y = \sum_{i=1}^{M_x} \Theta_i / \sum_{i=1}^{M_x} \varphi_i \quad (6)$$

The **second** step consists of expanding the bivariate density function as $n_{d,c_y} = n_d(d) \delta(c_y - \bar{c}_y)$ using a one-point quadrature centered at the solute mean concentration $\bar{c}_y$. Making use of this expansion, multiplying both sides of Eq.(1) by $v(d) c_y$ and integrating with respect to c_y from zero to $c_{y,max}$ and with respect to d from d_{min} to d_{max} one can get the mean solute concentration in the dispersed phase:

$$\frac{\partial[\bar{c}_y \phi_y]}{\partial t} + \frac{\partial}{\partial z} \left[\bar{c}_y \bar{F}(t, z) - D_y \frac{\partial[\bar{c}_y \phi_y]}{\partial z} \right] = \frac{Q_y^{in}}{A_c} \bar{c}_y^{in} \delta(z - z_y) + \bar{K}_{oy} (c_x^* - \bar{c}_y) \quad (7)$$

$$\bar{F}_y = \int_{d_{\min}}^{d_{\max}} u_y(\bar{c}_y, d) v(d) n(d, z, t) \partial d \approx \sum_{j=1}^{N_q} u_y(\bar{c}_y, d_j) v(d_j) w_j \quad (8)$$

$$\bar{K}_{oy} = \int_{d_{\min}}^{d_{\max}} \frac{6K_{oy}(d, \bar{c}_y)}{d} v(d) n(d, z, t) \partial d \approx \sum_{j=1}^{N_q} \frac{6K_{oy}(d_j, \bar{c}_y)}{d_j} v(d_j) w_j \quad (9)$$

where the unclosed integrals in the convective and mass transfer terms are evaluated using the QMOM based on the known moments of the marginal density $n(d, z, t)$, where the weights (w_j) and the abscissas (d_j) are found using the product-difference algorithm as illustrated by McGraw (1997). The solution for this unknown marginal density is accomplished by the generalized fixed-pivot technique using the system of Eqs.(4) and (5), where the required moments for the evaluation of the unclosed integrals could be accurately predicted. The accuracy of predicting at least two moments (the zero and third moments) is one property of the generalized fixed pivot technique (Attarakih et al., 2004b). The first and second moments are usually predicted with high accuracy that allows a two-point Gauss quadrature to evaluate the integrals appearing in Eqs.(8) and (9). Although a two- point quadrature is used in this work, a three- point quadrature could also be used since most of the higher moments of the known distribution, $n(d, z, t)$, could be accurately estimated.

4. Spatial coordinate discretization

Eqs.(4) and (5) or (7) through (9) represent a system of conservation laws that are coupled through the convective and source terms and are dominated by the convective term for typical values of D_d and U_d encountered in LLECs (*Peclet No.* $\approx 1 \times 10^3 H - 2 \times 10^3 H$). Due to the dominance of the convective term it is expected that the hold up profile of each class (ϕ_i) will move with time along the column height with a steep front. So, accurate front tracking discretization approaches are to be used such as the nonoscillatory first and second order central difference schemes. Let the i th convective flux be denoted as $F_i = U_{d,i} \phi_i$ and the staggering spatial grid: $z_{l \pm 1/2} = z_l \pm \Delta z / 2$ and the

average cell hold up as $\phi_{i,l} = \int_{z_{l-1/2}}^{z_{l+1/2}} \phi_i(t, z) \delta z / \Delta z$. The convective flux is then discretized in conservative form using the Kurganove and Tadmor (2000) central schemes (see Attarakih et al., 2004b), while the implicit Euler method is used for time discretization.

5. Numerical results and discussion

To completely specify the problem, the following geometry is used for a laboratory scale LLEC: $H=2.55$ m, $z_d=0.25$ m (the dispersed phase inlet), $z_c=2.25$ m (the continuous phase inlet), column diameter = 0.15 m, the inlet feed is normally distributed with mean droplet diameter of 3 mm and standard deviation of 0.5 mm. The inlet solute concentrations in the continuous and dispersed phases are taken as 50 and 0 kg/m³ respectively and the total flow rate of each phase is taken 2.778×10^{-5} m³/s. The terminal droplet velocity is evaluated from the Vignes (1965) correlation based on the procedure described by Gourdon et al. (1994) using water-acetone-toluene as a chemical system, where the direction of mass transfer is from the continuous to the dispersed

phase. The slowing factor and the droplet interaction functions are described in detail by Schmidt et al. (2004). The initial condition is taken as zero (no dispersed phase present initially). All the numerical testes are conducted using a PC processor of 1.4 MGH speed and Compaq Visual FORTRAN version 6.6.

Fig. 1 shows the solute concentrations in the dispersed and continuous phases at steady state using two-dimensional grid of size: 30×100 , where the droplet active mechanism is droplet breakage. It is clear that both profiles as predicted by the detailed and reduced models are almost identical. This is in fact elucidates the accuracy of the reduced model, and reveals clearly the efficiency of the generalized fixed-pivot technique for its ability to furnish the moments required for the inversion of the moment problem to get the weights and the abscissas required to evaluate Eqs.(8) and (9). Fig.(2) shows again the same concentration profiles as in Fig.(1); however, both droplet breakage and coalescence are now active. It is also clear how both models produced identical results; however, at the expense of the computational time.

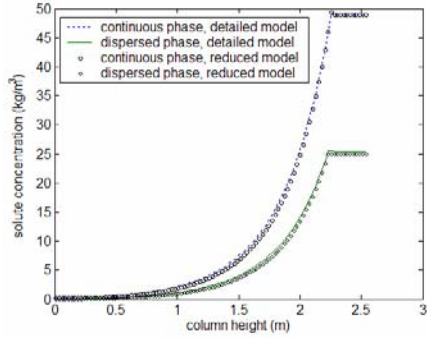


Figure1: Steady state solute concentration as predicted by the detailed and reduced models for droplet breakage only at 250 rpm.

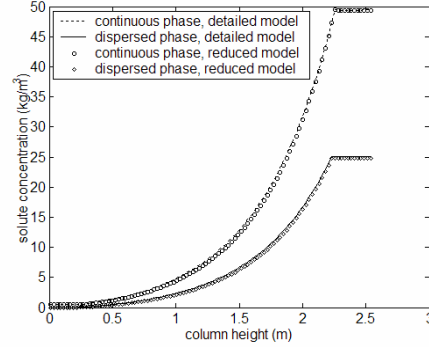


Figure2: Steady state solute concentration as predicted by the detailed and reduced models for droplet breakage and coalescence at 250 rpm.

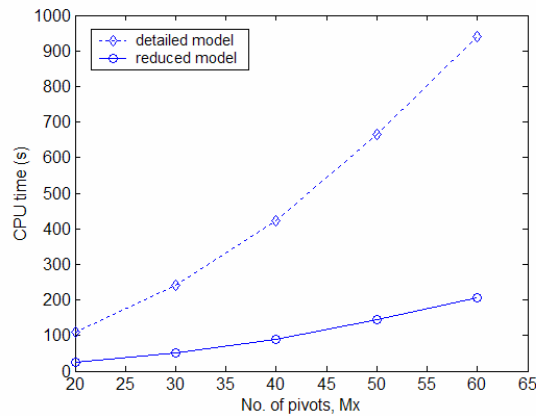


Figure 3: The CPU time requirements for the detailed and reduced models.

Fig.(3) shows the considerable reduction in the computational time when the reduced model is used. This is because the dimensionality of the problem is reduced from $2M_x+1$ to M_x+2 partial differential equations. The surprising accuracy of the reduced model is due the detailed information that is included in the reduced model without any simplifying assumptions regarding the shape of the distribution. The effect of the solute concentration on the column hydrodynamics and visa versa is discussed in detail by Attarakih et al. (2004a) and Schmidt et al. (2004).

6. Conclusions

- A comprehensive bivariate population balance model is presented to predict the behavior of the spatially distributed population balances for LLECs by coupling the hydrodynamics and mass transfer through the breakage, coalescence frequencies and the droplet velocity.
- The detailed and reduced models presented in this work are found to produce almost identical predictions for the coupled hydrodynamics and mass transfer. The application of the QMOM to the reduced model equations is found very effective in estimating the unclosed integrals.
- The reduced model shows a substantial reduction in the CPU time when compared to the detailed one without any loss of accuracy.

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