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A New Approach to Solve Population Balance Equations in Chemical Processes

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Abstract
Nowadays, population balance is used as a powerful engineering tool in design issues. Different processes in chemical, petrochemical, biotechnology, pharmaceutical industry, etc. deal with particles. In such processes particle or bubble size distribution (PSD) influences the final product quality and also process design. On the other hand solution to the dominant hydrodynamic and thermo-kinetic equations ignoring this distribution will make it impossible to accurately simulate these processes. Solution to population balance equations (PBE's) is needed to attain the PSD.
Since 1940's many researchers have proposed different methods for easier and also more accurate solution of PBE's. One of the most common and famous of these methods is the classes method (CM). However, as this method requires a large number of classes to give a reasonable result, it needs a huge amount of calculations and time. To overcome this problem, in this paper a new variant of CM is proposed in which particles in different classes are transformed to new classes in two steps. In the first step, the particles are agglomerated and broken up to form three parallel types of groups, namely: groups formed from agglomerated particles; groups formed from broken up particles and finally a group formed from non-altered particles. In the second step, these parallel groups are combined to redefine classes for next time step. Finally, results of this method, which could be called Parallel Groups Classes Method (PGCM), for different coalescence and breakage kernels are compared with those obtained using analytical solution and CM. Excellent agreement of results of PGCM with analytical solution reveals its effectiveness and accuracy; which will give it an advantage over CM.

Keywords: Population balance, Classes method, Particle size distribution, Agglomeration, Break-up.

Introduction

Population balance is used in several engineering applications. It is a well-established method used to analyze the particle size distribution (PSD) of the dispersed phase in processes such as precipitation, polymerization, crystallization, food processing, PSD of crashed raw material in mineral processes, rain drops and so on.
In many of these processes population balance equations (PBE's) should be coupled and solved simultaneously with fluid dynamic equations. In this case a huge amount of calculations should be done. As analytical solutions exist in very few cases [1], numerical techniques are sought. To all of these techniques accuracy and low time consumption are essential.
Since 1940's various techniques and methods have been proposed to ease the solution of PBE's. The earliest published studies were done by Blatz and Tobolsky [2] who

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considered the coagulation and fragmentation of a polymerization-depolymerization system.
Other famous methods are Monte Carlo Method [3], classes method (CM) [4-5], the standard method of moments (SMM) [6], the quadrature method of moments (QMOM) [7-9], the direct quadrature method of moments (DQMOM) [10, 11] and multiple size group method (MUSIG) [12].
In this paper a new approach is proposed to solve CM with less effort and more accuracy. As this method uses parallel independent groups of particles in each stage of size evolution, it is called parallel group classes method (PGCM).

Theory

A class of particles is defined a set of particles close in size so that it can be assumed they all have the same size called the characteristic size of the class. So if a dispersed phase consisting of particles with different sizes is to be divided to *M* classes, a set of characteristic sizes of classes is defined as $\{J_1, J_2, ..., J_M\}$ so that a particle which belongs to *ith* class has a size $x_i \in [J_{i-1}, J_i)$; for the first class $x_1 \in [0, J_1)$.
In some cases J_M can be so large to be assumed infinity; however, in many practical cases physical properties confine it to a limited value, such as size of a bubble which is limited by maximum stable bubble size [13].
A method to calculate the characteristic size of a class is the sauter mean diameter which is defined as:

$$d_{32,i} = \frac{\sum_{j=1}^{N_i} x_j^3 n_j}{\sum_{i=1}^{N_i} x_j^2 n_j}, \qquad (1)$$

where $d_{32,j}$ is the mean sauter diameter of *ith* class, N_i is the total number of particles in the *ith* class, and n_j is the number of particles having a diameter x_j (note that $\frac{p}{6} x_j^3 \in [J_{j-1}, J_j)$).
In addition to this, a characteristic size could be fixed (fixed pivot) i.e. once it has been calculated or specified it will remain unchanged throughout calculations, or it can be variable (moving pivot) i.e. in the beginning of each time step the characteristic size is recalculated from the results of the last time step.
Population balance equation in the simplest form can be written as:

$$\frac{\partial N_i}{\partial t} + \nabla.(\vec{u} N_i) = r_B - r_D, \qquad (2)$$

In which N_i is the number of particles in the *ith* class, $\vec{u}$ is the velocity vector of flow field, and r_B and r_D are the rate of birth and death of particles in that class, respectively.
Equation (2) is frequently stated with a distribution function, $f(v,t)$, which relates to N_i, the total number of particles within ith class, as:

$$N_i(t) = \int_{J_{i-1}}^{J_i} f(v,t) dv \qquad (3)$$

f can also be approximated by a finite set of Dirac delta functions as:

$$f(v,t) \cong \sum_{i=1}^{M} N_i(t) d(v - x_i) \quad (4)$$

Thus after summing up equation (2) and substitution of N_i with f a new form of PBE is acquired:

$$\frac{\partial f}{\partial t} + \nabla.(\vec{u} f) = R_{C,B} \quad (5)$$

where $R_{C,B}$ indicates rate of coalescence and breakage of different particles expressed as function of distribution function.

r_B and r_D in eq(2) are results of agglomeration (coalescence), breakage, nucleation, reaction, absorption and dissolution phenomena. Ignoring the last four processes which have small or none contribution in some chemical operations (other than crystallization), these two terms may be written as follows:

$$r_B = r_{B,a} + r_{B,b}, \quad (6)$$

$$r_D = r_{D,a} + r_{D,b}, \quad (7)$$

where indices *a* and *b* correspond to agglomeration and breakage, respectively.

Moreover if the convective term can be neglected (in fact this is not the case in reality, however, many authors have done so to verify their technique and compare it with available analytical solutions) the original PBE can be written as:

$$\frac{\partial N_i}{\partial t} = r_{B,a} - r_{D,a} + r_{B,b} - r_{D,b}. \quad (8)$$

Now the above rates should be defined. In terms of the continuous distribution function, *f*:

$$r_{B,a} = \frac{1}{2}\int_0^v Q(v, v-u) f(v,t) f(v-u,t) du, \quad (9)$$

$$r_{D,a} = f(v,t)\int_0^{\infty} Q(v,u) f(u,t) du, \quad (10)$$

$$r_{B,b} = \int_v^{\infty} g(u) b(u) p(u \mid v) f(u,t) du, \quad (11)$$

$$r_{D,b} = b(v) f(v,t). \quad (12)$$

where Q and b are the coalescence and breakage rates respectively, g is the number of particles formed from a breakage and $p(u \mid v)$ is the fraction of particles of size v that have been formed from breakage of a parent particle of size u.

In terms of the discrete variable N_i, the following equations can be easily derived by substitution of f from equation (4) into Eq's (9)-(12). So:

$$r_{B,a} = \frac{1}{2}\sum_{j=1}^{i-1} Q(x_j, x_{i-j}) N_j N_{i-j} \quad \text{where} \quad x_j + x_{i-j} \in [J_{i-1}, J_i), \quad (13)$$

$$r_{D,a} = N_i \sum_{j=1}^{M} Q(x_i, x_j) N_j, \quad (14)$$

$$r_{B,b} = \sum_{j=i}^{M} g(x_j) b(x_j) p(x_j \mid x_i) N_j, \quad (15)$$

$$r_{D,b} = b(x_i) N_i. \quad (16)$$

The formulation of new method starts with assuming M original classes in the beginning of time step t^n. Three types of parallel independent groups are formed after coalescence and breakage phenomena, which are:

a) M group consisting of particles formed from breakage of original particles. So if a particle is formed after break-up of a particle in the original *ith* class, its size will be denoted by $z_k^{(i)} \in [J_k^{(i)}, J_{k+1}^{(i)}) \subseteq [0, x_i)$ and the number of particles in breakage classes by $B_k^{(i)}$ with $k = 1,..., NB_{(i)}$.

b) $M(M+1)/2$ groups consisting of particles formed from agglomeration of original particles. So if a particle is made up from coalescence of a particle originally from *ith* class and another particle from *jth* class with $i, j = 1,...,M$. Then its size and number are denoted by $y^{(ij)} (= x_i + x_j \in [J_{i+j-2}, J_{i+j}))$ and A_{ij}, respectively.

c) non-altered particles which form a group of original classes and have noting as the same as original classes, except that the total number of particles in ith class is shown with $\overline{N}_i$.

In this regard the number of particles in *ith* class can be written as a sum:

$$N_i = \overline{N}_i + A_{ij} + B_k^{(i)}. \tag{17}$$

And the PBE can be split into three equations:

$$\frac{d\overline{N}_i}{dt} = -r_{D,a} - r_{D,b}, \tag{18}$$

$$\frac{dA_{ij}}{dt} = r_{B,a}, \tag{19}$$

$$\frac{dB_k^{(i)}}{dt} = r_{B,b}. \tag{20}$$

As a result the original PBE is transformed into 3 simpler equations. These equations are solved to give separate size distributions which in turn are combined to form the overall size distribution in the new time step.

To start simulation one needs to specify kernels and an initial condition. Herein it's done as the following and according to Scott [1].

Coalescence kernels: Agglomeration kernels were selected as:

1. Constant kernel: $Q(u,v) = C$
2. Summation kernel: $Q(u,v) = b(u+v)$
3. Product kernel: $Q(u,v) = Buv$

where $Q(u,v)dt$ is the coalescence probability per unit densities in the ranges du and dv, and *C*, *b* and *B* are constants. For the sake of simplicity and in order not to lose the available analytical solutions, the system is assumed with zero particle breakage.

Dimensionless parameters are defined as $X = J/J_0$ or x/J_0, $f(X,T) = J_0 f(J,t)/N_0$ and T is a dimensionless time which is defined in each case. For a constant kernel $T = CN_0 t$, for summation kernel $T = bN_0 J_0 t$, and for the product kernel $T = BN_0 J_0^2 t$.

Initial conditions: A continuous Gaussian-like distribution approximated by a function of the following form was considered as the initial condition:

$$f(v,0) = \frac{N_0 a^{n+1}}{v'\Gamma(n+1)} \left(\frac{v}{v'}\right)^n e^{-av/v'}, \tag{21}$$

where *a*, v' are constants and *n* is a positive number.

It can easily be shown that for this type of initial distribution, the number of particles having size of $5J_0$ is less than 1 percent of total number of particles (J_0 is initial mean size of all particles which can be found to be $J_0 = v'(n+1)/a$). So as no breakage of particles is considered, in a short time from start-up of process it is reasonable to assume a maximum particle size of $20J_0$.

Of the aforementioned kernels the former and the latter are chosen. For constant agglomeration kernels, the dimentionless analytical distribution function is derived to be [1]:

$$f(X,T) = \frac{(1-t)\exp\{-(t+n+1)X + (n+2)Xt^{1/(n+2)}\}}{X^{3/2}t^{(2n+3)/(2n+4)}[2p(n+2)/(n+1)]^{1/2}} \times \left[1 - \frac{(2n+3)(n+3)}{24Xt^{1/(n+2)}(n+1)(n+2)}\right] \quad (22)$$

where $t = 1 - e^{-T}$. And for product kernel it is:

$$f(X,T) = \frac{(n+2)\exp\{-X(T+n+1) + X(n+3)T^{1/(n+3)}[(n+1)/(n+2)]^{(n+2)/(n+3)}\}}{X^{3/2}[T(n+2)/(n+1)]^{(2n+5)/(2n+6)}[2p(n+1)/(n+3)]^{1/2}} \times \left[1 - \frac{(n+4)(2n+5)}{24XT^{1/(n+3)}(n+1)^{(n+2)/(n+3)}(n+2)^{1/(n+3)}(n+3)}\right] \quad (23)$$

Results

The new PGCM has been validated on aggregation and breakage processes. A comparison of PGCM with CM and analytical solutions are presented.

Figures 1 and 2 show total number distribution and the distribution function relative errors respectively, calculated for a constant coalescence kernel by applying CM to particles having a maximum size of $10J_0$ and $20J_0$ ($X_{max} = 10$ and 20 respectively) and lying in 10 and 20 classes.

From Fig 1 one can conclude as number of classes reduces the total number of particles in each class increases. It also shows that there is still a significant difference between analytical solution and the results obtained from CM. Fig 2 shows that the error increases with size of class but considering more classes will decrease this error considerably.

Fig 3 shows a comparison between analytical solution and results of 20 and 10 classes obtained from PGCM. There is no significant difference although there is a great reduction in number of classes. Moreover a good consistency of results is observed with analytical results. Note that for both of these two curves $X_{max}/M = 1$. Otherwise the comparison would become meaningless.

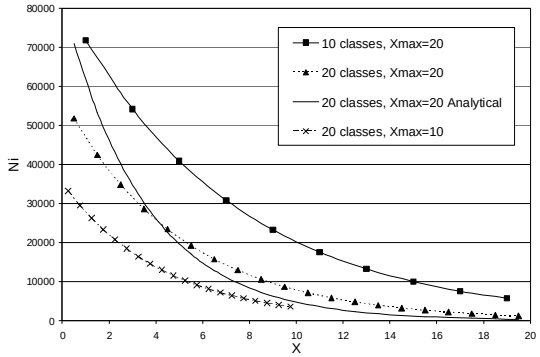

Figure 1. Total number of each class versus dimensionless size for T=5.

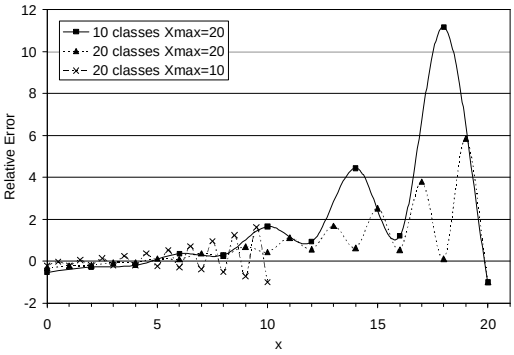

Figure 2. Relative distribution function error comparing to analytical solution for T=5.

Fig 4 shows a similar plot to that of fig 3 but with a product agglomeration kernel. It also shows that PGCM of 10 classes and of 20 classes (here noted as PGCM-10 and PGCM-20) give much better results than CM-20.

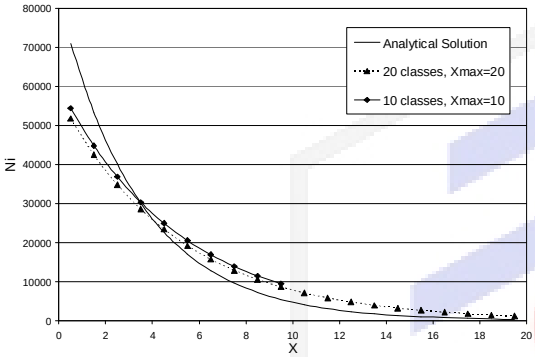

Figure 3. Comparison of analytical solution and PGCM with 10 and 20 classes for a constant agglomeration kernel.

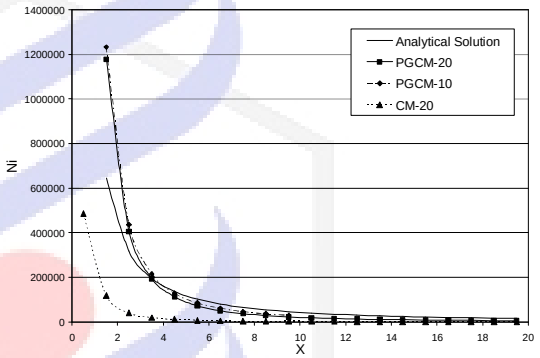

Figure 4. Comparison of analytical solution with PGCM of 10 and 20 classes and CM of 20 classes.

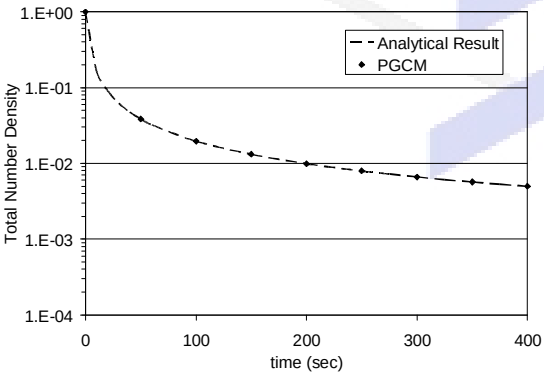

Figure 5. Comparison of analytical solution and PGCM with 10 classes for a constant agglomeration kernel.

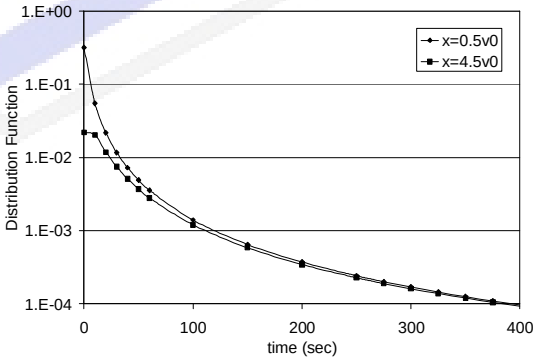

Figure 6. Distribution functions of 2 classes for a constant agglomeration kernel obtained from PGCM with 10 classes.

Fig 5 shows a comparison of analytical solution and PGCM-10 for variation of total number of particles versus time. The good agreement is clear. In figure 6 the distribution functions for two classes of $0.5J_0$ and $4.5J_0$ are shown. In figures 3-8 other data are shown including distribution functions, classes evolution and also comparison between PGCM and CM.

Conclusion

In this article a novel technique for solving population balance equations has been proposed. The new approach is formulated to enhance the classes method. The method has good accuracy in comparison with the original CM but with fewer defined classes. It predicts moments of PSD very well. For more accuracy the number of breakage classes should be increased. The numerical simulations have shown the ability of PGCM to predict the evolution of moments and main properties of the population balance of an asymptotic PSD by using few classes with a good accuracy. The plots of relative errors have revealed that PGCM is able to predict a large range of fractional moments with small errors. From a CFD viewpoint the new technique has a great advantage over others, as it needs surely lower computational time.
In the future works, PGCM will be applied to predict a more complex system namely a bubble column. It is anticipated that this technique shows its advantages in this case as well.

Literature Cited

[1] Scott W.T., "Analytic Studies of Cloud Droplet Coalescence I", *Int. J. Atmosph. Sci.*, **25** (1967), 54.
[2] Blatz P.J., Tobolsky A.V., "Note on the Kinetics of Systems Manifesting Simultaneous Polymerization-Depolymerization Phenomena", *J. Phys. Chem*., **49** (1945) 77.
[3] Vikhansky A., Kraft M., "A Monte Carlo method for identification and sensitivity analysis of coagulation processes", *J. Comp. Phys.*, **200** (2004) 50.
[4] Vanni M., "Approximate population balance equations for aggregation-breakage processes", *J. Colloid Interface Sci.*, **221** (2000) 143.
[5] Ramkrishna D., "Population Balances", Academic Press, San Diego, 2000.
[6] Gimenez M., Clausse A., "An application of the method of moments to the modeling of bubbly flow", *Annals of Nuclear Energy*, **32,** (2005) 1705.
[7] McGraw R., "Description of aerosol dynamics by the quadrature method of moments", *Aerosol Sci. Technol.*, **27** (1997) 255.
[8] Gordon R.G., "Error bounds in equilibrium statistical mechanics", *J. Math. Phys.*, **9** (1968) 655.
[9] McGraw R., Wright D.L., "Chemically resolved aerosol dynamics for internal mixtures by the quadrature method of moments", *J. Aerosol Sci.*, **34** (2003) 189.
[10] Fan R.D., Marchisio D.L., Fox R.O., "Application of direct quadrature method of moments to poly-dispersed gas-solid fluidized beds", *J. Aerosol Sci.*, **139** (2004) 7.
[11] Marchisio D.L., Fox R.O., "Solution of population balance equations using the direct quadrature method of moments", *J. Aerosol Sci.*, **36** (2005) 43.
[12] Lo, S., "Application of population balance to CFD modeling of gas-liquid reactors", *Proceedings of Conference on Trends in Numerical and Physical Modeling for Industrial Multiphase Flows, Corse, 27-29 September 2000.*
[13] Clift R., Grace J.R., Weber M.E., "Bubbles, Drops and Particles", 1978, Academic Press, New York.