ANALYSIS OF POPULATION BALANCE—IV†

THE PRECISE CONNECTION BETWEEN MONTE CARLO SIMULATION AND POPULATION BALANCES

DORAISWAMI RAMKRISHNA

School of Chemical Engineering, Purdue University, West Lafayette, IN 47907, U.S.A.

(Received 6 February 1980; accepted 20 November 1980)

Abstract—Dispersed phase systems may be analyzed via population balances or by Monte Carlo simulation methods. This paper establishes the precise mathematical connection between the two procedures. The population balance equation is obtained by averaging the master density equation, while simulation procedures efficiently evaluate the solution to the master density equation by elimination of low probability events.

INTRODUCTION

Dispersed phase systems occur so commonly in chemical engineering that population balances, the natural implement for their analysis, belong as a standard component of the chemical engineer's analytical repertoire. The general framework of population balances appeared first in the chemical engineering literature with the publication of Hulburt and Katz[1]. Subsequently, their statistical origin together with the augmented machinery for small populations in which random fluctuations would be important, were presented by Ramkrishna and Borwanker[2,3]. The author has discussed the general features of population balances in regard to their applicability to chemical engineering problems[4]. Randolph and Larson[5] discuss population balances with particular reference to crystallization systems.

Briefly, population balances arise in the synthesis of information about single particle behavior into a description of the behavior of an entire population of such particles. The equations which result are frequently integrodifferential because conservation of the number of particles of a specific state must account, for example, for their creation arising from the cumulative contribution of particles of other states. Such integrodifferential equations have been solved for scalar particle state variables.

An alternative approach to predict the behavior of a population from models for single particle behavior has been via Monte Carlo simulation methods. Thus Spielman and Levenspiel[5] have used simulation methods to investigate the effects of drop-mixing on reaction conversion in the dispersed phase droplets of a liquid-liquid system. Zeitlin and Tavlarides[6] have simulated the behavior of stirred liquid-liquid dispersions to predict the stabilized drop size distribution in an agitated vessel. Shah et al.[7] have presented a general simulation procedure for particulate systems based on the concept of an interval of quiescence first employed in an early work on biological populations by Kendall[8]. The foregoing procedure has been employed by the author and his coworkers in numerous situations[9-14]. Gupta et al.[15] have used the method for simulating the behavior of solid-liquid suspensions under the effect of shear.

Simulation methods are attractive for several reasons. Firstly, they obviate the need for solving integrodifferential population balance equations, which are especially difficult for multivariate number density functions. Secondly, the simulation results can produce not only average values but also provide information about fluctuations characteristic of small populations. Thirdly, when interaction occurs between two or more particles (such as in an agglomerating population) correlations may develop between particle states which are not accounted for in calculations based on population balances. (See for example Ramkrishna et al.[10]). This is due to the fact that the population balance equation, normally employed, assumes the particle states to be statistically independent. Such situations are more directly investigated by simulation methods.

The objective of this work is to inquire into the precise connection between simulation methods and the framework of population balances. Such a connection exists since both consist in synthesizing information about single particle behavior into that pertaining to the population. More specifically, it is of interest to see the mathematical connection between the two apparently different routes to the same answers. Furthermore, simulation methods are capable of generating information on fluctuations about average behavior, a feature not in general possessed by the population balance equation. In this connection, it would then also be our objective to relate the expanded framework of product density analysis (see Ramkrishna and Borwanker[2,3]), that can account for fluctuations about mean behavior, to simulation methods. Before proceeding to meet these objectives, a brief discussion of simulation methods is presented.

SIMULATION METHODS

The system of interest to us is a population of particles distinguished from each other by a set of say n quantities selected such that their specification for a particular particle determines the behavior of that particle. By behavior we imply two possible processes; one refers to changes in particle state involving no loss of identity of the particle. Such changes could result for example from rate processes such as heat and/or mass transfer. Even
motion through physical space is an example of change of physical location. We assume such changes to be deterministic (to which extent our analysis is limited) and given by ordinary differential equations which arise from a model of single particle behavior. Thus such changes result in smooth variations in the particle state. The second process refers to those that destroy the identity of a given particle. Examples of these are the breakage of a particle into smaller particles or agglomeration of a particle with other particles. Processes of this type lead to a change in the total number of particles. More abstractly, we have the situation of particles distributed in an n-dimensional continuum through which they are continuously in motion with "velocities" depending on their instantaneous locations. Furthermore, particles are created and destroyed at rates given by source and sink terms respectively. This abstract picture will be extremely helpful in subsequent discussions.

A simulation method applied to a physical system, in which random changes occur, is in essence an artificial realization of the system. This artificial realization must of course be consistent with the statistical laws which specifically apply to the system of interest. Thus a simulation algorithm is normally established by identifying the probability functions which govern changes occurring in the system. The implementation of the algorithm depends on generating random numbers satisfying the calculated probability distributions. Thus the generation of random numbers provides a prescription for the change to be introduced into the system. Indeed the probability distributions for the changes to be introduced are conditional on the state of the system at the stage at which the change is considered. For a process such changes occur along time and progressive simulation produces a particular realization of the process. The average behavior of the system is obtained by averaging the various "sample paths" of the system by repeated simulations.

We should now be a little more specific. In simulation a particulate system described at the outset it is clear that the smooth changes in the particle states occurring during the period in which they retain their identity are obtained by solving differential equations governing the change of particle state. What is uncertain however is the length of time over which the particle remains intact. This has been referred to as the quiescence interval in which they retain their identity are obtained by solving differential equations governing the change of particle state. The probability that the population at time t consists of Y particles, with volume in which the particles are located about x_i, i = 1, 2, ..., Y is given by

\[ J_2(x_1, x_2, \ldots, x_Y; t) \, dV_1 \, dV_2 \ldots dV_Y \]  

If V represents the volume in which the particles are constrained, the above master density function has the normalization condition

\[ \sum_{i=1}^{Y} \frac{1}{i!} \int_V dV \int_V dV \ldots \int_V dV J_i(x_1, x_2, \ldots, x_i; t) = 1. \]  

The division by \( i! \) in eqn (2) is to account for the fact that particles of identical states are indistinguishable (see [2] for more details). The average of any quantity over the population is calculated by including it in the inte-
Analysis of population balance—IV

In order to derive the Janossy density equation, we must consider the mutually exclusive and exhaustive ways under which a stipulated situation at time $t+\Delta t$, may be attained from conditions at time $t$. Thus suppose that at time $t+\Delta t$, there are $\nu$ particles in the system with sizes between $x_i$ and $x_i+dx_i$, $i=1,2,\ldots,\nu$. The probability of this occurring is

$$J_\nu(x_1, x_2, \ldots, x_\nu; t+\Delta t) dx_1 dx_2 \ldots dx_\nu$$

(5)

This situation may arise by either of two possible ways. There may exist at time $t$, $\nu$ particles of sizes between $(x_i-dx_i)$ and $(x_i-dx_i+dx_i)$, $i=1,2,\ldots,\nu$ and during the time interval $t$ to $t+\Delta t$ none of the particles break. The probability of this occurring is

$$J_\nu(x_1 - dx_1, x_2 - dx_2, \ldots, x_\nu - dx_\nu; t)$$

(6)

The differential $dx_i'$ must satisfy the condition

$$dx_i' = \Delta X(x_i - dx_i') dt$$

(7)

so that the above particles may grow to the required sizes during the interval $dt$.

The second way in which the envisaged situation at time $t+\Delta t$ may arise is by any one of the following mutually exclusive ways; a typical case is that there are at time $t$, $(\nu-2)$ particles with sizes between $(x_i-dx_i)$ and $(x_i-dx_i+dx_i)$, $i=1,2,\ldots,\nu$ but excluding say $j$ and $k$ and a $(\nu-1)$st particle of size between $(x_j+x_k-dx')$ and $(x_j+x_k-dx'+dx_j$) which breaks during the time interval into two particles, one of size $x_j$ and the other of size $x_k$. The probability of this occurring is given by

$$\sum_{j=1}^{\nu} \sum_{k=1}^{\nu} J_{\nu-1}(x_1 - dx_1, \ldots, x_{j-1} - dx_{j-1}, x_j + x_k - dx', x_{j+1} - dx_{j+1}, \ldots, x_{k-1} - dx_{k-1}, x_k + dx', x_{k+1} - dx_{k+1}, \ldots, x_{\nu} - dx_\nu; t) \Gamma(x_j + x_k - dx')$$

Equating expression (5) to the sum of (6) and (8), dividing by $dt$ and letting $dt \to 0$, we obtain the light of (7)

$$\frac{\partial}{\partial t} J_\nu + \sum_{j=1}^{\nu} \frac{\partial}{\partial x_j} \left[ \Delta X(x_j) J_\nu \right] = -\sum_{j=1}^{\nu} \Gamma(x_j) J_\nu$$

(9)

For a more general derivation of eqn (9), in which particle state is considered a vector, see Ramkrishna[17]. Equation (9) must be subject to an initial condition.

**The Connection with Simulation Methods**

There are basically two options available for calculating $f_\nu(x; t)$ starting from eqn (9). Consistent with eqn (3),
we may directly average eqn (9) to obtain the population balance eqn (4). To this end, we consider \( x \) in eqn (9) to be \( x \), integrate the equation w.r.t. \( x_1, x_2, \ldots, x_{\nu-1} \) between the limits \( 0 \) and \( \infty \), divide by \( (\nu - 1)! \) and sum over all \( \nu \). Using (3), we have

\[
\frac{\partial}{\partial t} f(x; t) + \frac{\partial}{\partial x} \left[ \bar{X}(x)f(x; t) \right] + \sum_{\nu} \frac{1}{(\nu - 1)!} \left\{ \prod_{k=1}^{\nu-1} \int_0^\infty dx_k \bar{X}(x_k)\bar{J}_\nu \right\} = -\Gamma(x)f(x; t)
\]

\[
- \sum_{\nu} \frac{1}{(\nu - 1)!} \prod_{k=1}^{\nu-1} \int_0^\infty dx_k \bar{X}(x_k)\bar{J}_\nu + \sum_{\nu} \frac{1}{(\nu - 1)!} \prod_{k=1}^{\nu-1} \int_0^\infty dx_k \bar{X}(x_k)\bar{J}_\nu
\]

\[
2 \int_0^\infty dx \sum_{\nu} \frac{1}{(\nu - 1)!} \prod_{k=1}^{\nu-1} \int_0^\infty dx_k \bar{X}(x_k)\bar{J}_\nu
\]

\[
J_{\nu-1}(x_1, \ldots, x_{\nu-1}, x_{\nu+1}, \ldots, x_n; t)\Gamma(x)f(x; t)
\]

We now explore the second option of computing \( f(x; t) \) from (3) which means that the Janossy density function (9) must be solved first. Indeed, the solution must be considered w.r.t. some \textit{initial condition}. Suppose now that the initial state of the population is prescribed by the initial value of \( f\), such as

\[
f(x; 0) = \phi(x).
\]

The expression \( r = 1, i \) is used in eqn (10) to denote the exclusion of \( r = i \). In simplifying eqn (10), we first recognize that \( \bar{X}(x)\bar{J}_\nu \) must vanish at the boundary \( x = 0 \) and \( x = \infty \). Further, in dealing with the third term on the r.h.s. of (10), we let \( x_i = x_i + x \), and replace the integration variable \( x_i \) by \( x_i \). Since it is readily perceived that

\[
\int_0^\infty dx_1 \int_0^\infty dx_2 \ldots = \int_0^\infty dx_1 \int_0^\infty dx_2 \ldots
\]

the normalization condition

\[
\int_0^\infty p(x'_n, x_k) \, dx_k = 1
\]

(expressing the fact that the size of a piece broken off a particle cannot exceed the size of the latter), together with the fact \( p(x'; x) = p(x', x' - x) \) convert the third term of the r.h.s. of (10) into

\[
\sum_{\nu} \frac{1}{(\nu - 1)!} \prod_{k=1}^{\nu-1} \int_0^\infty dx_k J_{\nu-1}(x_1, \ldots, x_{\nu-1}, x_{\nu+1}, \ldots, x_n, x; t)\Gamma(x)
\]

\[
J_{\nu-1}(x_1, \ldots, x_{\nu-1}, x_{\nu+1}, \ldots, x_n, x; t)\Gamma(x)
\]

where we have renamed \( x_i \) as \( x_i \) for convenience. The symmetry of \( J_{\nu-1} \) makes each of the integrals in the innermost summation (on index \( k \)) equal. Since there are \( (\nu - 1) \) such integrals, the expression (12) becomes

\[
\sum_{\nu} \frac{1}{(\nu - 2)!} \prod_{k=1}^{\nu-2} \int_0^\infty dx_k J_{\nu-2}(x_1, \ldots, x_{\nu-2}, x; t)\Gamma(x)
\]

which obtains on renaming of the coordinates. On replacing \( (\nu - 1) \) by \( \nu \), expression (13) is identical to the second expression on the r.h.s. of (10) with the sign reversed. Thus the third term cancels the second term. The fourth term becomes

\[
2 \int_0^\infty dx' \sum_{\nu} \frac{1}{(\nu - 1)!} \prod_{k=1}^{\nu-1} \int_0^\infty dx_k \bar{X}(x_k)\bar{J}_\nu
\]

\[
J_{\nu}(x_1, \ldots, x_{\nu-1}, x', x_{\nu+1}, \ldots, x_n; t)\Gamma(x')p(x', x)
\]

so that eqn (10) leads exactly to the population balance equation (4) as one may have anticipated. Clearly, therefore \( f_0(x; t) \) could be obtained by solving the population balance equation. We will not be concerned with this procedure any further here.

We now explore the second option of computing \( f(x; t) \) from (3) which means that the Janossy density equation (9) must be solved first. Indeed, the solution must be considered w.r.t. some \textit{initial condition}. Suppose now that the initial state of the population is prescribed by the initial value of \( f_0 \), such as

\[
f_0(x; 0) = \phi(x).
\]

Let the total number of particles at \( t = 0 \) be given by

\[
N_0 = \int_0^\infty \phi(x) \, dx.
\]

Although \( N_0 \) is only the expected value, we may assert that there are \textit{exactly} \( N_0 \) particles since the detail with which the initial state of the population is known does not necessitate a more probabilistic description. In practice even \( N_0 \) may not be known for it is more usual to state the initial size distribution of the particles, in which case the choice of \( N_0 \) becomes flexible. In order to calculate the Janossy density function \( J_{\nu}(x_1, x_3, \ldots, x_n; t) \), we must solve eqn (9) subject to an initial value at \( t = 0 \). However, the solution for some intermediate interval should be adequate to represent the evolution of the system from the very beginning. The Janossy density function \( J_{\nu}(x_1, x_3, \ldots, x_n; t) \) may be regarded as conditional on the \textit{initial state} of the population, although the notation does not explicitly indicate so.

We now consider the solution of eqn (9) subject to some initial condition at \( t' < t \). This solution may be accomplished by the method of characteristics (see [19]). Notice that eqn (9) features only the densities \( J_{\nu-1} \) and \( J_{\nu} \), reflecting how the population may increase by only one at a time. Hence in the time interval \( t' \) to \( t \) we may have at most an increment of one particle. If however the interval \( t' \) to \( t \) is broken up into \( n \) sub-intervals by locating \( (n - 1) \) intermediate times, an increment of \( n \) particles could be considered by solving eqn (9) successively in each of these intervals starting from the earliest interval. Of course a countless number of partitions exist.
of the interval \((t', t)\) into \(n\) sub-intervals and an \((n - 1)\)-fold iterated integration is called for to account for this combinatorial complexity. It is this combinatorial complexity that makes it difficult to make practical use of a solution of eqn (9). In view of the foregoing discussion, it should be clear that a solution of eqn (9) for the interval \((t', t)\) should be sufficient to establish the entire solution based on a stepwise marching strategy.

Equation (9) must be integrated along the characteristic curve in \(v\)-dimensional space. In the evolution of the system from \(t'\) to \(t\) we are concerned with either of two alternatives. The first is that there were only as many \((v)\) particles at \(t'\) as there are at \(t\) so that none of the particles underwent breakage during the interval \((t', t)\). In this case the characteristic curve is in \(v\)-dimensional space throughout the time interval \((t', t)\) originating at \(t'\) at a point obtained by solving the differential equation

\[
\frac{dx}{dt} = \hat{X}(x), \quad u < t
\]  

(18)

\[
x(x) = x_{n}, \quad j = 1, 2, \ldots, v.
\]

(19)

We denote the solution of (18) subject to (19) as

\[
X(x, t|t'), \quad j = 1, 2, \ldots, v
\]

(20)

which is the characteristic curve with its latest location at \((x_1, x_2, \ldots, x_v)\) and going backwards to that at \(t'\). Clearly \(X(x, t|t') = x\). The second possibility is that the \(v\) particles at \(t\) arise from \((v - 1)\) particles at time \(t'\) and one of the particles break at some instant \(t', \, (t' < t < t)\). The characteristic curve in this case is confined to the fixed instant \(t'\), \((t' < t < t)\) and continues in \(v\)-dimensional space during the time interval \((t', t)\). Thus there is a discontinuity in the characteristic curve in this situation.

To sum up the foregoing discussion the solution of eqn (9) must be obtained along the characteristic curve, which is a smooth continuous piece if there is no particle breakage in the interval \((t', t)\), or has a single discontinuity at \(t'\) where a single breakage event occurs. Figure 1 shows these two situations well. The system consist of initially one particle at point \(A\) and the growth of this particle along AB until at \(B\) it splits into two particles located at \(C\). The two-particle system evolves in 2-dimensional space along the section \(CD\), which represents the growth of both particles until at \(D\) one of the two particles splits into two making the system jump to \(E\) in a 3-dimensional domain. The characteristic curve \(EF\) represents the evolution of the three-particle system.

The integration of eqn (9) to calculate \(J_\cdot\) at time \(t\), given its value at \(t'\), is now conveniently expressed as

\[
J_\cdot(x_1, x_2, \ldots, x_v; t) = J_\cdot(X(x_1, t|t'), X(x_2, t|t'), \ldots, X(x_v, t|t'), \prod_{j=1}^{v} \hat{X}(X(x_j, t'|t''))) \hat{X}(x)
\]

(21)

The ratio of growth rate terms occur because of the distortion of particle size intervals with time due to growth. For vectorial particle states, these will be replaced by appropriate Jacobians.

\[
Pr[T > \tau|\text{State of population at } t'] = \exp \left[ -\sum_{j=1}^{n} \int_{t'}^{t} \Gamma(X(x_j, t|u)) du \right]
\]

(22)

represents the probability that, given there are \(v\) particles at time \(t'\) with sizes \(X(x_1, t|t'), \ldots, X(x_v, t|t')\), none of the particles break up during the time interval \((t', t)\). This interval is referred to as being quiescent. It is convenient to define a quiescence interval \(T\) as the time elapsed since \(t'\) during which none of the \(v\) particles present at time \(t'\) with sizes, say \(x_i\), \(i = 1, 2, \ldots, v\), break up and precisely one of the particles break up at \(t + \tau\). Then

\[
Pr[T > \tau|\text{State of population at } t'] = \exp \left[ -\sum_{j=1}^{n} \int_{t'}^{t} \Gamma(X(x_j, t|u)) du \right]
\]

(23)

where \(X(x_i + u|x_i, t')\) is used to denote the size of the particle at time \(t' + u\), given its size is \(x_i\) at \(t'\) and is obtained by integrating forwards eqn (18). Note that (22) and (23) are essentially the same if it is recognized that in
(22) the focus on particle size is that at \( t \) (which is specified) whereas in (23) it is on the size at \( t' \). The probability density of quiescence interval distribution conditional on the state of the population at \( t' \) is given by

\[
\frac{1}{\sigma} \frac{\partial}{\partial \tau} P(x_1, x_2, \ldots, x_n; t') = \sum_{j=1}^{n} \Gamma(X(t' + \tau|x_1, t')) \exp \left[ -\sum_{j=1}^{n} \int_0^{\infty} \Gamma(X(t' + \tau|x_1, t')) \, du \right].
\]

(24)

It is now possible to see from (21) how the simulation technique of Shah et al.[7] is arrived at. The second expression on the r.h.s. of (21) contains \( J_{-1} \), in turn must satisfy a relation similar to (21). Successive substitution will produce on the r.h.s. of (21), besides others, the following term

\[
\sum_{i=1}^{\infty} \sum_{j=1}^{n} \int_0^{t'} \int_0^{t} J_{-1}(X(x_1, t|t'), \ldots, X(x_{i-1}, t|t'), X(x_i, t'|t'), \ldots, X(x_n, t|t'))
\]

\[
\left[ \sum_{i=1}^{\infty} \sum_{j=1}^{n} \Gamma(X(x_i, t|t')) \right] \exp \left[ -\int_0^{t'} \left[ \sum_{i=1}^{\infty} \sum_{j=1}^{n} \Gamma(X(x_i, t|t')) \right] \, du \right]
\]

(25)

where we have used \( x_i = X(x_1, t|t') \) to represent the size of the particle which breaks at \( t' \). Expression (25) represents the mutually exclusive and exhaustive ways in which one breakage event between \( t' \) and \( t \) can produce the situation envisaged at \( t \) starting from compatible conditions at \( t' \). More specifically, the quiescence interval (conditional on the state of the population at \( t' \) as appearing in the argument of \( J_{-1} \), \( t'' - t' \) is allowed to vary from 0 to \( (t - t') \) in (25) in the integration w.r.t. \( t' \); note particularly that, in view of (24), the second term in the main integrand is the quiescence interval distribution. The third term in (25) consists of

\[
\left[ \sum_{i=1}^{\infty} \sum_{j=1}^{n} \Gamma(X(x_i, t|t')) + \Gamma(x_1) \right]
\]

(26)

which represents the probability that, given that the quiescence interval is \( t'' - t' \), the particle which breaks at \( t'' \) is the \( j \)th particle of size \( x_j \), and

\[
p(x_j, X(x_k, t|t''))
\]

(27)

is the probability density function which identifies the sizes of product of breakage as those which become \( x_j \) and \( x_k \) in subsequent growth (without further breakage) at time \( t \). Expressions (24), (26) and (27) clearly bring out the simulation strategy of Shah et al.[7]. Given the number of particles at \( t' \) and their sizes, (24) lays down the distribution function according to which random numbers representing the quiescence interval must be generated. Expression (26) shows the rule for random number generation, which identifies the particular particle which breaks at \( t'' \), while (27) affixes the sizes of broken particles.

An important conclusion to make therefore is that the averaging of sample paths from the foregoing simulation procedure is in effect evaluating the average from (3). A schematic representation of the various options for calculation of the mean quantities and their fluctuations associated with the population is presented in Fig. 2.

In closing, it is useful to observe that the average quiescence interval for the density given by (24) is calculated readily when \( \Gamma \) is independent of size. Thus if \( \Gamma(x) = \gamma \), a constant, (24) becomes \( \gamma e^{-\gamma t} \) so that the average quiescence interval is \( 1/\gamma \), which could be used as a unit of discretization of the time interval for a more

---

**Fig. 2. Population balances vs simulation procedures.**
simplified simulation procedure analogous to that of Spielman and Levenspiel[5] or Zeitlin and Tavlarides[6]. Finally, it should be obvious that the development in this paper could have been done in an entirely identical manner for agglomerating populations or more generally for populations in which both breakage and agglomeration events occur. The generalization to systems in which reciprocal changes occur between the population and its environment (continuous phase) is also done in a relatively straightforward manner.

CONCLUSIONS

We have established precisely how simulative methods for the behavior of particulate systems and population balances are connected in the synthesis of single particle behavior into population behavior. The solution of the Janossy density equation, obtained by successive substitutions displays the tremendous combinatorial complexity that is characteristic of the temporal evolution of a particulate system. Monte Carlo simulation methods are in essence an efficient way of obtaining numbers from such combinatorially complex analytical solutions by eliminating relatively lower probability events by artificial realization.

NOTATION

- expectation
- expected population density or product density of order one
- Janossy or master density
- actual number density per unit volume of particle state space; also dimension of particle state space
- probability density for the size distribution of particles formed by breakage of a parent particle
- distribution function for quiescence interval
- time
- volume in particle state space
- variable particle size
- variable state vector
- size of given particle
- particle growth rate

Greek symbols

- transition probability function for particle breakage
- initial condition for expected population density

REFERENCES