Analytical Solution of Population Balance Equation Involving Growth, Nucleation and Aggregation in Terms of Auxiliary Equation Method

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Abstract: The Auxiliary Equation Method (AEM) has been modified to obtain the solutions of a Population Balance Equation (PBE) involving particulate growth, nucleation and aggregation phenomena. In all the cases examined, the volume density distributions are accurately predicted by the travelling wave solutions of the complementary equation of the nonlinear partial integro-differential equation with distinctly chosen parameters. Being a flexible technique and a direct comparison for the existing analytical solutions, this study proves the potential of the proposed methodology.

Keywords: Population Balance, Growth, Nucleation, Aggregation, Auxiliary Equation method, Travelling wave.

1 Introduction

Particulate processes are characterized by size distributions that are assumed to vary strongly in time with respect to mean particle size and shape of the Particle Size Distribution (PSD). Population balances are widely encountered in numerous scientific and engineering disciplines to describe the evolution of PSD in processes that involve particulate phenomena like growth, nucleation, aggregation and breakage [1]. These particulate phenomena may occur during the process which may result in momentous changes manifested through its PSD. The time evolution of this distribution is determined by the solution of the so-called Population Balance Equation (PBE) which governs the dynamic behavior of particulate processes through a nonlinear partial integro-differential equation within the mathematical framework [1]. The numerical solution of a dynamic PBE is a remarkably complicated problem due to both numerical complications and the uncertainties of the model regarding the particulate mechanisms that are frequently weakly approximated. On the other hand, the essence of any numerical solutions of a PBE requires the discretization of the particle diameter/volume domain by means of certain numerical approximations that results in a system of stiff, nonlinear integro-differential equations.

Since the early 1960s, various numerical methods have been developed for solving PBM involving both time-dependent and -independent formulations. Among those include the fully discrete method [2,3], method of classes [4,5], fixed and moving pivot method [6,7], higher order discretized method [8,9], orthogonal collocation on finite elements [10,11], Galerkin and wavelet-Galerkin method [12,13], Monte Carlo [14] and least squares method [15,16]. Several other techniques available for solving the PBEs are the various method of moments where the PBE is solved for the moments of the PSD, e.g., quadrature method of moments (QMOM) [17,18] and direct quadrature method of moments (DQMOM) [19,20].

Despite numerous papers published on the numerical solutions of a PBE, the choice of the most appropriate method for the calculation of time evaluation of a PSD, in particular processes undergoing simultaneous particle
growth, nucleation, breakage and aggregation, is not straightforward and realistic. In reality, a large number of studies refer to only a limited range of variation of particle breakage and aggregation rates. Therefore, the wide-ranging application of a numerical method to the solution of a specific problem cannot be assured. On the other hand, the formulation of a large number of different numerical approaches also underlines the intrinsic problems in obtaining a precise and consistent numerical method. In this study, a one-dimensional (1D) population balance equation (PBE) under various conditions of particulate nucleation, growth and aggregation are investigated and compared with their analytical and numerical solutions as found in the literature. Then the analytical method introduced by Pınar and Özis [21,22] is implemented to solve the PBE and the validity of the solution is established through example case studies involving growth, nucleation and aggregation processes.

2 Problem Formulation

Population balance modeling has become an important tool to model a wide variety of particulate processes and its increasing applicability in various disciplines of science and engineering. For example, processes like crystallization, granulation, milling, polymerization, flocculation, and aerosols involve population balances. The PBE describes the evolution of a density function, representing the behavior of a population of a state vector such as size or volume of solid particles, liquid droplets or gas bubbles. The rapid development in this field is possible thanks to the availability of improved particle size measurement techniques to measure multivariate distributions. The evolution of this density function takes into account the different processes that control the population such as aggregation, breakage, growth and advective transport of the state vector. The generalized one-dimensional Population Balance Equation (PBE) in a well-mixed control volume, without considering spatial dependence, is written as:

$$\frac{\partial}{\partial t} n(x,t) + \frac{\partial}{\partial x} [G(x,t)n(x,t)] = A_{\text{nuc}} + A_{\text{agg}} + A_{\text{break}} \quad (1)$$

where

$$A_{\text{nuc}} = B_0(t)\delta(x_0)$$

$$A_{\text{agg}} = \int_0^\infty \! n(x-x',t)n(x',t)a(x-x',x')dx'$$

$$- \int_0^\infty \! n(x,t)n(x',t)a(x,x')dx' \quad (2)$$

$$A_{\text{break}} = \int_x^\infty \! b(x,x')n(x',t)\Gamma(x')dx' - \Gamma(x)n(x,t)$$

where the $n(x,t)$ is the number density function in terms of the particle volume $x$. $a(x,x')$ is the volume-based aggregation kernel that describes the frequency at which particles with volume $x$ and $x'$ collide to form a particle of volume $x + x'$. $\Gamma(x)$ is the volume-based breakage function and the stoichiometric kernel $b(x + x')$, satisfying the symmetry and normalization conditions, gives the product size distribution for binary breakage through the probability of formation of particles with volume $x$ from the breakage of particles of volume $x'$.

3 The Auxiliary Equation Method (AEM) approach

The Auxiliary Equation method (AEM) suggested recently by Pınar and Öziş [21,22] has been applied to nonlinear physical models and has successfully helped to develop new analytical solutions with appropriate parameters and these parameters are chosen such a way that the obtained solutions simulates the behavior of the solutions of aforementioned problems. Recently, Pınar et al [23] proposed analytical solutions of PBEs involving particulate aggregation and breakage using AEM and compared the results with their available analytical solutions obtained from the literature for a 1D PBE. In this study, the proposed methodology is further extended to include growth and nucleation within the mathematical framework. The detailed features of this method can be read from the recommended papers of Pınar and Öziş [21,22]. For the sake of brevity, the adjustment of the analytical method for the proposed solution is briefly mentioned here and the detail is left for the interested readers to refer to Pınar and Öziş [21,22] and Pınar et al [23].

In the previous section, a particulate population balance is introduced briefly. In this study we shall develop an analytical particle distribution conjecture which works well in the case of certain well-defined standard processes of particle formation. The distribution function is defined by Eq. (1) and the physical parameters affecting the formation of the distribution are nested in this differential equation. In our conjecture, we adapt the previously mentioned works of Pınar and Öziş [21,22] to determine the solution of the batch PBE problem. Our conjecture is based on the reinterpretation of particle phase space and amalgamates it with the methodology as suggested in Pınar and Öziş [21,22]. Particle phase space consists of least number of independent coordinates attached to a particular distribution that allow a complete description of the properties of the distribution. Particle phase space and may conveniently be divided into two sub regions given by internal and external particle coordinates. External coordinates refer simply to the spatial distribution of the particles. Such external coordinates of course are not necessary, for example, in the description of a well-mixed particulate process, although it may be quite convenient to report the
distribution on a unit value basis. Internal coordinate refers to those properties that are attached to each particle that quantitatively measure its state, independent of its position. A common example of an internal coordinate property is particle volume. The particle volume distribution is defined so that \( n(x,t)dx \) is the number of particles per vessel volume at time \( t \) in the volume range \((x,x+dx)\) and therefore the total volume \( \int_0^\infty n(x,t)dx \) must also be conserved [24]. In any particulate process giving rise to the formation of PSD, individual particles are continuously changing their position in the particle phase space, namely, each particle moves along the various internal and external coordinate axes. If these changes are gradual and continuous, one refers to this movement as convection along the respective particle coordinates and refers to the rate of change of the coordinate property of a particle as the convective particle velocity along that coordinate axes [24].

Hence, in this study, the particle velocity is conjectured and assume to be slow enough and continuous and the particle velocity components are nested in the function \( \zeta = \zeta(x,t) \) where \( x \) the particle volume and time \( t \). Because, internal convection velocity can be taken as linear rate of growth of a particle and \( \zeta = \zeta(x,t) \) can be chosen a linear combination of particle volume \( x \) and the time \( t \). An alternative to tracking the convective particle velocity is to fix it by a suitable choice of new space coordinates i.e. \( \zeta = \zeta(x,t) \) and solve the problem in this new coordinate system. As such, following the methodology in Pınar and Öziş [21,22] the nonlinear partial integro-differential equation i.e. the batch PBE (Eq. 1) can be readily reduced to ordinary integro-differential equation using an independent single variable \( \zeta \) where \( \zeta = \zeta(x,t) \) is a function of particle volume \( x \) and time \( t \). Consequently Eq. (1) and the complementary equation are invariant under the appropriate transformation \( \zeta = \zeta(x,t) \) and also their solutions.

To solve ordinary integro-differential equation, by using the solution ansatz (see Eq. (3) in Pınar and Öziş [21]) and balancing the ansatz, one can easily determine the solution series as a second order polynomial

\[
v(\zeta) = g_0 + g_1z(\zeta) + g_2z^2(\zeta) \tag{3}
\]

In \( z(\zeta) \) and the coefficients \( g_0, g_1 \) and \( g_2 \) the free parameters can be further determined. Referring to the initial condition (i.e. initial value) which is in the form of an exponential distribution in the PBE model, the expected solution is therefore always exponential. Referring to Case 6 of Table 1 in Pınar and Öziş [21], the auxiliary equation can be expressed as:

\[
\left( \frac{dz}{d\zeta} \right)^2 = a_2z^2(\zeta) + a_6b(\zeta) \tag{4}
\]

with the solution

\[
z(\zeta) = e^{-\frac{1}{2}LambertW \left( -\frac{\ln \exp(\frac{\sqrt{\Phi}}{2\phi})}{2\phi^2} \right) - \sqrt{\phi^2(\Phi - \phi)}} \tag{5}
\]

which behaves in an exponential form. Using the methodology in Pınar and Öziş [21,22], the parametric general solution of the PBE is given for various case studies of growth, nucleation and aggregation processes and compared to their analytical solutions that exist in the literature [25,26,27].

4 Case study 1: Combined Nucleation, Growth and Aggregation

The population balance for a combination of nucleation, growth and aggregation case can be obtained from Eq. 1 by setting the breakage function and the specific rate of breakage to zero i.e. \( \hat{b}(x,x') = 0, \Gamma(x) = 0 \). This type of PBE is typically encountered in MSMPR systems [28,29] and in crystal synthesis [30,31]. For the sake of comparison with an analytical solution, the growth and nucleation kernels for the steady-state PBE are assumed to be constants i.e. \( G(x,t) = G_0 = 1, B_0(t) = \beta_0 = 1, a(x,x') = 1 \). Using the aforementioned conditions, Liao & Hulburt [32] proposed the following analytical solution:

\[
n(n) = 2n_0 \exp(-px) I_1(x) \frac{t}{x} \tag{6}
\]

where

\[
p = \sqrt{1 + \frac{1}{2\beta_0 n_0 G_0 t^2}}
\]

\[
x = \frac{\sqrt{2 \beta_0 n_0 G_0}}{G_0}
\]

\[
n(0) = n_0
\]

Using the proposed AEM approach, the analytical solution obtained is as follows:

\[
n(x,t) = g_0 + g_1e^{-\frac{1}{2}LambertW \left( -\frac{\ln \exp(\frac{\sqrt{\Phi}}{2\phi})}{2\phi^2} \right) - \sqrt{\phi^2(\Phi - \phi)}} + g_2e^{-\frac{1}{2}LambertW \left( -\frac{\ln \exp(\frac{\sqrt{\Phi}}{2\phi})}{2\phi^2} \right) + \sqrt{\phi^2(\Phi - \phi)}}
\]
where

\[ \Phi = -\mu x + \alpha x + \mathcal{C} I \]

\[ g_0 = -10\sqrt{e^{-100x}} \]

\[ g_1 = -10\sqrt{2e^{(x-0.1)}} - 2.2 \]

\[ a_2 = \frac{2000\sqrt{e^{-100x}}}{\alpha \mu} \]

\[ a_6 = -0.01\tanh(0.1x) \]

\[ \mu = 1.5, \alpha = -0.3, \mathcal{C} I = -0.3 \]

Further in this case, the nucleation function is also set to zero i.e. \( B_0(t) = 0 \). In classical modelling approach, the growth and aggregation phenomena are independently considered. However, applications occur \cite{33} where the growth and the aggregation are coupled thus not obeying the classical Von Smoluchowski equation. Till date, only a few analytical solutions have been proposed in literature for various combinations of growth and aggregation kernel functions as seen in Table 1.

![Graph showing comparison](image)

**Fig. 1:** Comparison of the proposed AEM analytical solution with the analytical solution of Liao & Hulburt \cite{27} (Case study 1)

As seen in Figure 1, there is a general similarity in the behavior between the proposed AEM solution and the analytical solution proposed by Liao & Hulburt \cite{27}. However, the slight difference in the solutions is anticipated. This is because even if both the solutions are inherently analytical, the postulations of the derivation of these solutions differ from one other. As Liao & Hulburt \cite{27} mentioned in their work, the combined aggregation, growth and nucleation was obtained by setting the breakage function and specific rate of breakage to zero and additionally the aggregation kernel and growth function were both taken to be constants. However, in our case, they are supposed to vary with the leading term volume \( x \).

### 5 Case study 2: Growth and Aggregation

Similar to the previous case, the PBE for a combination of growth and aggregation case is obtained by setting the breakage function and the specific rate of breakage to zero. The corresponding analytical solution using AEM approach is obtained as follows:

\[
n(x,t) = \frac{\frac{M_0^2}{M_1}}{1 - 2\Lambda x_0 \left( \frac{N_0 - M_0}{M_0} \right)} \exp \left[ -\frac{M_0^2}{M_1} x - 2\Lambda x_0 \left( \frac{N_0 - M_0}{M_0} \right) \right] \tag{7}
\]

**Table 1**: Combinations of growth and aggregation kernel functions (Case study 2) obtained from Majumder et al \cite{34} and as found in Ramabhadran et al. \cite{26}.

<table>
<thead>
<tr>
<th>Case</th>
<th>( G(x,t) )</th>
<th>( a(x,x') )</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>1</td>
<td>100</td>
</tr>
<tr>
<td>b</td>
<td>( x )</td>
<td>( x + x' )</td>
</tr>
</tbody>
</table>

According to case 2a (see Table 1), the analytical solution given by Ramabhadran et al. \cite{26} is given as follows:

\[
n(x,t) = \frac{\frac{M_0^2}{M_1}}{1 - 2\Lambda x_0 \left( \frac{N_0 - M_0}{M_0} \right)} \exp \left[ -\frac{M_0^2}{M_1} x - 2\Lambda x_0 \left( \frac{N_0 - M_0}{M_0} \right) \right] \tag{7}
\]

where

\[ M_0 = \frac{2N_0}{2 + \beta_0 N_0 t} \]

\[ M_1 = N_0 x_0 \left[ 1 - \frac{2G_0}{\beta_0 N_0 x_0} \ln \left( \frac{2}{2 + \beta_0 N_0 t} \right) \right]. \]

Here \( \Lambda = \frac{G_0}{\beta_0 N_0 x_0} \) and \( M_0 \) and \( M_1 \) are the moments.
where
\[ g_0 = 10e^{-0.2x^2}, \quad g_1 = -e^{-x}, \quad g_2 = -e^{-x}, \quad a_6 = -10e^{-x}, \quad a_2 = \frac{\tanh\left(\frac{x}{1000}\right)}{1000} \]
\[ \Phi = -\mu x + \alpha + CI, \quad \mu = 1.4, \quad \alpha = -0.5, \quad CI = 0.0 \]

A comparison of the two analytical solutions can be seen in Figure 2.

Note that \( I_1 \) is the modified Bessel function of first kind of order one.

The corresponding analytical solution using AEM approach is obtained as follows:
\[ n(x,t) = \frac{1}{3} + g_1 e^{-\frac{1}{2} \text{LambertW}\left(-\frac{\Phi(x)}{\mu^2} + \sqrt{\Phi} \right)} + 6g_1^2 e^{-\frac{1}{2} \text{LambertW}\left(-\frac{\Phi(x)}{\mu^2} + \sqrt{\Phi} \right)} \frac{1}{18x + 1} \]

where
\[ g_1 = 10e^{-7.84x^2}, \quad a_2 = -10e^{-x}, \quad a_6 = 25\tanh(1000) \]
\[ \Phi = -\mu x - \frac{6g_1^2(18x + 1)\mu}{g_1^2} \]
\[ \mu = 2.8, \quad \alpha = 3.5, \quad CI = -4.0 \]

A comparison of the two analytical solutions can be seen in Figure 3.

It is worth noting that the AEM solutions of cases 2a and 2b, similar to case 1, preserve the general trend of the exponential distribution and the slight difference is again based on the derivation of the analytical solutions based on distinct assumptions.
6 Case study 3: Nucleation and Growth

Similar to the previous cases, the PBE for a combination of nucleation and growth is obtained by setting the breakage function and the specific rate of breakage to zero. However in this case, instead of the nucleation function, the aggregation kernel is set to zero i.e. $a(x,x') = 0$. Studies related to combined nucleation and growth is undergoing a growing importance in nanotechnology [35]. Several analytical solutions have been proposed in literature for various combinations of nucleation and growth phenomena. From these, two cases with combinations of growth and nucleation kernel functions (see Table 2) are considered.

Table 2: Combinations of growth and nucleation kernels (Case study 3) as obtained from Kumar & Ramkrishna [25].

<table>
<thead>
<tr>
<th>Case</th>
<th>$G(x,t)$</th>
<th>$B_0(t)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>1</td>
<td>$10^3$</td>
</tr>
<tr>
<td>b</td>
<td>1</td>
<td>10</td>
</tr>
</tbody>
</table>

According to case 3a (see Table 2), Kumar & Ramkrishna [25] proposed the following analytical solution:

$$n(x,t) = n_0(x - G(t)) + \frac{N_{0,n}}{\sigma_0} \left[ \exp \left( -\frac{x_{low}}{x_{0,n}} \right) - \exp \left( -\frac{x}{x_{0,n}} \right) \right]$$

(10)

where $x_{low} = \max(x_1, x - \sigma_0 t)$.

In Eq. (10), $n_0(x)$ is the initial number density and $N_{0,n}$ and $\sigma_0$ indicate parameters related to exponentially distributed nucleation rate and growth rate function respectively.

The corresponding analytical solution using AEM approach is obtained as follows:

$$n(x,t) = g_0 + g_1 e^{-\frac{1}{2} \text{LambertW}(-\frac{\text{B}(t)}{\sigma_x^2} e^{\frac{1}{\sigma_x^2}(\Phi(x))}) + \sqrt{\Phi(x)}}$$

$$+ g_2 e^{-\frac{1}{2} \text{LambertW}(-\frac{\text{B}(t)}{\sigma_x^2} e^{\frac{1}{\sigma_x^2}(\Phi(x))}) + \sqrt{\Phi(x)}} e^{\Phi(x)}$$

(12)

where

$$g_0 = 10 e^{-(x+0.6)^2}$$

$$g_1 = -e^{-x-0.02}$$

$$g_2 = -e^{-x-0.02}$$

$$a_2 = \frac{\tanh(x + 0.02)}{1000}$$

$$a_6 = -10 e^{-x-0.02}$$

$$\Phi = -\mu x + \alpha + CL$$

$$\mu = 1.6, \alpha = -0.8, CL = -3.0$$

According to case 3b (see Table 2), Kumar & Ramkrishna [25] proposed the following analytical solution:

$$n(x,t) = n_0(xe^{-G_0 t}) e^{-G_0 t} + \frac{N_{0,n}}{\sigma_0} \left[ \exp \left( -\frac{x_{low}}{x_{0,n}} \right) - \exp \left( -\frac{x}{x_{0,n}} \right) \right]$$

(11)

where $x_{low} = \max(x_1, xe^{-G_0 t})$

The corresponding analytical solution using AEM approach is obtained as follows:

$$n(x,t) = g_0 + g_1 e^{-\frac{1}{2} \text{LambertW}(-\frac{\text{B}(t)}{\sigma_x^2} e^{\frac{1}{\sigma_x^2}(\Phi(x))}) + \sqrt{\Phi(x)}}$$

$$+ g_2 e^{-\frac{1}{2} \text{LambertW}(-\frac{\text{B}(t)}{\sigma_x^2} e^{\frac{1}{\sigma_x^2}(\Phi(x))}) + \sqrt{\Phi(x)}} e^{\Phi(x)}$$

(12)

where

$$g_0 = 10 e^{-(x+0.6)^2}$$

$$g_1 = -e^{-x-0.02}$$

$$g_2 = -e^{-x-0.02}$$

$$a_2 = \frac{\tanh(x + 0.02)}{1000}$$

$$a_6 = -10 e^{-x-0.02}$$

$$\Phi = -\mu x + \alpha + CL$$

$$\mu = 1.6, \alpha = -0.8, CL = -3.0$$

The proposed AEM solutions related to cases 3a and 3b are compared with their corresponding available
analytical solutions, as seen in Figures 4 and 5 respectively. As can be clearly observed, the solutions proposed using AEM and suggested by Kumar & Ramkrishna [25] match very well. The slight differences that exist between the solutions are again based on the derivation of the analytical solutions based on distinct assumptions. In the literature, several analytical formulations are available to represent various particulate processes based on various hypothesis and/or assumptions. But, in principle, such analytical formulations should be equivalent and can essentially be transformed from one to the other through certain equations. Although this proposal is theoretically correct, but in practice this is not observable in many cases because the population density versus size function typically used to model particulate processes and the population, in general, is used as the distributed variable rather than mass/or particle volume. For example, in several PBM’s linear size is used as an independent variable rather than particle volume for simplicity. But in particulate processes, when aggregation (agglomeration) or breakage is more important the particle volume must be used rather than size (say, diameter). It is important to note that although the volume (or mass) is conserved in these processes, linear size is not conserved. Figure 6 shows the volume conservation behavior of this approach using case 2a (as an example). \( \mu \) is the volume coefficient as we use transformation which depends on both volume and time. As seen in Figure 5, when \( \mu \) changes the curve does not change thus indicating volume (mass) conservation. The AEM solution proposed in this study is found to be compatible with the various analytical solutions obtained from the literature, and thus adds to the belief that AEM is a more robust approach.

7 Conclusions

In this study, an effective analytical technique is implemented to solve PBEs involving simultaneous particulate growth, nucleation and aggregation by making use of the appropriate solution(s) of associated complementary equation via auxiliary equations. Travelling wave solutions of the complementary equation of the nonlinear partial integro-differential equation with appropriately chosen parameters is taken to be analogous to the description of the dynamic behavior of the particulate processes of a PBE. Using carefully chosen parameters, the AEM solution is able to reproduce the expected behavior of various particulate conditions and is compatible with the analytical solutions proposed in the literature.

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