ADCHEM 2006

International Symposium on Advanced Control of Chemical Processes
Gramado, Brazil – April 2-5, 2006

CHALLENGES OF MODELLING A POPULATION BALANCE USING WAVELET

Johan Utomo, Nicoleta Balliu, Moses O. Tadé[1]

Department of Chemical Engineering, Curtin University of Technology,
GPO Box U 1987, Perth, WA 6845, Australia.

Abstract: Crystallization is one of the oldest separation technologies due to its ability to produce a range of bulk products to high purity chemicals. Aspect of controlling the size distribution is important for downstream operations and characteristics of products. Two cases of population balance problems are considered in this paper to show limitations of some utilized methods. Those cases present the sharp transition phenomena in the particle size distribution. A wavelet-based method by Liu and Cameron (2001) is applied and compared with other conventional methods based on Finite Difference, Orthogonal Collocation and Orthogonal Collocation with Finite Elements. The result show that the wavelet method is faster, more accurate and more efficient in solving the population balance problems.

Keywords: Crystallization, Modelling, Population Balance, Wavelet method

1. BACKGROUND

Crystallization is one of the oldest separation technologies and plays a key role regarding the quality of the products and the economy of a whole plant. This process is used to manufacture large quantities of bulk materials as well as high purity chemicals.

Although crystallization technology has been established for a long time it is difficult to operate and control. Controlling particle size distribution (PSD), shape distribution and crystal purity are challenging due to the complexity and non-linearity of the process and because of lack of reliable on-line instrumentation to measure the key parameters (Rohani, et al. 1999; Braatz 2002). These properties affect downstream operation such as filtration, washing, drying, mixing and formulation (Braatz 2002; Fujiwara, et al. 2005). They also affect the end-usage properties such as entrainment liquid after dewatering, dissolution rate for pharmaceutical products, caking properties, fluidization properties, pneumatic handling properties, bulk density, and esthetic appearance (Randolph and Larson 1988).

According to Braatz (2002), inadequate control of particle size and shape can result in unacceptably long filtration or drying time, or in extra processing steps, such as re-crystallization or milling process. Shekunov et al. (2000) claim that in the pharmaceutical industry, there were advance control systems over drug identity and purity, however control over the physical properties such as form and crystallinity remains inferior. It is indicated that there are many attractive challenges arising from the pharmaceutical processes which can be used for further research directions especially in modelling and building advanced control systems.

Hulburt and Katz (1964) introduced a modelling approach for particulate processes more than 41 years ago. This approach is well known as the concept of population balances. The population balance equation (PBE) can be defined as a mathematical description characterizing particles undergoing the mechanisms of birth, growth, death and leaving a certain particle phase space. In

[1]Corresponding author : Tel: +61-8-9266-4998; Fax: +61-8-9266-2681; M.Tade@exchange.curtin.edu.au

crystallization, those mechanisms can be categorized as nucleation, growth, agglomeration and breakage.

A very sharp transition profiles problem commonly occurs in many chemical engineering cases. For example, concentration profiles in chromatography processes, temperature and activity profiles in solid catalyst, the profiles of reaction in fixed bed reactors as well as the particle size distribution for a well-mixed batch crystallizer in which crystal breakage and agglomeration may be neglected. These processes are represented by parabolic partial differential equations. Effective solutions for these models require certain type of numerical methods to be implemented.

2. NUMERICAL METHODS

2.1 Previous Methods

Most papers addressing population balance problems discussed techniques to solve systems from the unidimensional to multidimensional population balance models. Many numerical methods have been proposed such as method of moment, method of self-preserving distributions, method of weighted residuals, sectional method, and the discretization methods. Other methods, which have been used to solve PB problems, are also based on Monte Carlo method and finite element method. To sum up, the above methods can be categorized into four types i.e. finite difference approach, spectral methods (e.g. orthogonal collocation), finite element and other approach. There are major drawbacks from those methods such as high computationally cost, lack of stability and accuracy of the solution and the in applicability of the solved models for implementation in control based models. Extensive discussion of those methods can be found in the literature (Kostoglou and Karabelas 1994; Ramkrishna 2000; Vanni 2000). In this paper, simulation studies will be conducted to compare the computational efficiency, the accuracy as well as the stability between the finite difference method (FD), the orthogonal collocation (OC), the orthogonal collocation with finite element (OCFE) and the wavelet-based method.

2.2 Finite Difference Methods

Finite difference methods have been commonly used for the solution of all types of partial differential equations (ODEs) systems. FD method approximates the continuous function $f(x)$ with Taylor expansion series (Hangos and Cameron, 2001). They can be a first order or second order approximations. In our case, FD method is used to approximate the first partial derivative of population density over its size ($\partial n/\partial x$) and converts the PDE into a set of ODEs.

2.3 Orthogonal Collocation

This technique was developed more than 70 years ago and applied in various cases of boundary value problems. The trial functions are chosen as sets of orthogonal polynomials and the collocation points are the roots of these polynomials. The solution can be calculated from the collocation points. The use of orthogonal polynomials is to reduce the error as the polynomial order increases (Gupta 1995; Hangos and Cameron 2001).

2.4 Orthogonal Collocation with Finite Elements

The combination of dividing the regions into a number of elements and by applying orthogonal collocation techniques for each element can improve the solution where the profile is very steep. In the region where there is a sharp transition, numbers of small elements can be applied while the remainder utilizes larger size of elements. Selection of the elements size is therefore essential.

2.5 Wavelet-based method

In 2001, Liu and Cameron proposed wavelet based method to solve population balance problems. They developed Wavelet Orthogonal Collocation (WOC) and Adaptive Wavelet Orthogonal Collocation (AWOC) to solve agglomeration in batch vessel. Further information about wavelet method can be found in Liu's papers (Liu and Cameron 2001; Liu and Cameron 2003; Liu and Tade 2004). To our knowledge, wavelet method combined with Galerkin method was first applied in chemical engineering area by Chen et al. (1996) to solve the breakage mechanism in a batch crystallizer.

Significant advantages of using wavelet method are the accuracy in producing solutions in the sharp transition regions, computationally efficient solutions, stable and easily implemented solution that is applicable to another system. These advantages are related to the characteristic of wavelet method such as, localization properties in space and scale, hierarchical organization, sparse coefficients and easy handling of the derivatives as well as non-linear and integral terms. However in Liu and Cameron (2001), there was no comparative study between wavelet method and any other methods.

2.6 Daubechies orthonormal wavelets

Wavelet can be used as a basis function to represent a certain function. In the wavelet function, two-basis functions can be found, the scaling function and the wavelet function. The scaling function coefficient illustrates a local average of the function (coarse illustration) and the wavelet function coefficient describes detailed information of the function (refinements) that cannot be found from the average coefficient. Compared to Fourier expansion, wavelet approximation give smaller error and is highly localized at discontinuity regions (Nielsen 1998). Compared to the traditional trigonometric basis functions which have infinite support, wavelets have compact support, therefore wavelets are able to approximate a function by the placement of the right wavelets at appropriate locations. From Daubechies's

work (1988), scaling function (ϕ) and wavelet function (ψ) can be described by a set of L (an even integer) coefficients (p_k : k = 0,1,…, L-1) through the two-scale relationship:

$$\phi(x)=\sum_{k=0}^{L-1} p_k\,\phi(2x-k) \tag{1}$$

and the wavelet function

$$\psi(x)=\sum_{k=2-L}^{1}(-1)^k\,p_{1-k}\,\phi(2x-k) \tag{2}$$

The support for the scaling function is in the interval 0 to (L-1), whilst for the wavelet function is in the interval (1-L/2) to (L/2). The coefficients p_k are called the wavelet filter coefficients.

Denote $L^2(R)$ as the space of square integrable functions on the real line. Let V_j be the subspace as the L^2-closure of the linear combination of:

$$\phi_{jk}(x)=2^{j/2}\phi(2^j x-k) \tag{3}$$

for $k\in Z=\{..,-1,0,1...\}$. A function $f(x)\in V_j$ can be represented by the wavelet series:

$$f(x)=\sum_{k\in Z} f_{jk}\phi_{jk}(x) \tag{4}$$

The multi-resolution properties of wavelets give another advantage to represent functions in differential equations which can be solved numerically (Motard and Joseph 1994). Detailed information about Daubechies orthonormal wavelets can be found in Daubechies (1988).

2.7 Wavelet Orthogonal Collocation(WOC)

This method was proposed by Betoluzza and Naldi (1996) for solving partial differential equations. In 2001 it was developed and applied for solving population balance problems by Liu and Cameron (2001). The interpolation functions are generated by autocorrelation of the usual compactly supported Daubechies scaling functions $\phi(x)$. Then the function θ called autocorrelation function verifies the interpolation property due to the orthonormality.

$$\theta(0)=\int\phi(x)\phi(x)dx=1 \tag{5}$$

and

$$\theta(n)=\int\phi(x)\,\phi(x-n)dx=0,\ n\neq 0 \tag{6}$$

The approximate solution of our problem will be a function u_j in the term of its dyadic points to obtain the wavelet expression:

$$u_j(x)=\sum u_j(2^{-j}n)\,\theta(2^j x-n) \tag{7}$$

Detailed information can be found in Liu and Cameron (2001, 2003) and Bertoluza and Naldi (1996).

3. CASE STUDIES

We consider two case studies of population balance which have sharp and dramatic transition phenomena in their particle size distribution in the batch crystallizer. Even though the case studies considered here are simple, since the analytical solutions are available for comparison purposes, the more complex models can be solved using the methods described above.

3.1 Case I: Nucleation and size-independent growth

The population balance for nucleation mechanism and size independent growth is described by the partial differential equation:

$$\frac{\partial n(L,t)}{\partial t}+G\frac{\partial n(L,t)}{\partial L}=B_0 \tag{8}$$

where n is number of particle (population density), L is dimensionless particle size, $L\in[0,2]$, G is the growth rate ($G=1$) and B_o is the nucleation rate, $B_0=\exp(-L)$. The initial condition is $n(L,0)=0$ and the boundary condition when $L=0$, $n(0,t)=0$. The analytical solution for this case is:

$$\begin{aligned} n(L,t)&=1-\exp(-L) && ;L-t<0\\ n(L,t)&=\exp(-L)[\exp(-t)-1] && ;L-t>0 \end{aligned} \tag{9}$$

3.2 Case II: Size-independent growth only

One dimensional population balance for size dependent growth mechanism only is described by the partial differential equation:

$$\frac{\partial n(L,t)}{\partial t}+G\frac{\partial n(L,t)}{\partial L}=0 \tag{10}$$

with:

$$n(0,t)=0;\quad n(L,0)=\exp(-100(L-1)^2) \tag{11}$$

The independent growth rate ($G=1$) is constant. The range of dimensionless particle size, $L\in[0,4]$. The analytical solution for the second case is :

$$n(L,t)=\exp(-((L-G.t-1)\times 10)^2) \tag{12}$$

4. DISCUSSION

All the simulation results presented have been executed on a 3.00 GHz Pentium IV – 1.00 Gigabytes of RAM running under Windows 2000. A MATLAB® version 7.0.1 was used as the computation software to simulate the models.

4.1 Case I: Nucleation and size-independent growth

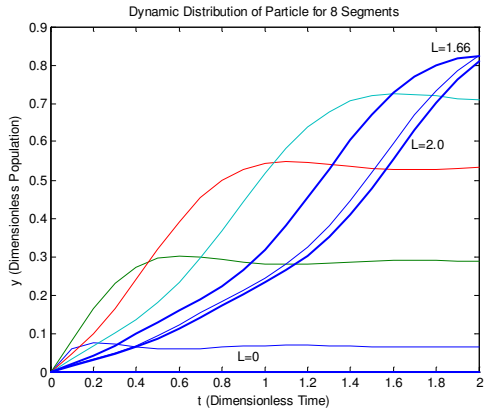

Fig.1. Dynamic distribution of 8 segments of particles in case I

Firstly we divide the size interval range into 8 segments and simulate the dynamic distribution of particles. It is seen from Figure 1 that using $L = 0$ the smallest segment of particles remains zero, while the other increase by following the underdamped mechanism until the overdamped mechanism for the largest segment of particles ($L = 2$). We found that the dynamic particle distribution gives the stable responses.

Finite difference method is employed to solve this problem numerically. The results are shown below in Figure 2 by using 101 discretization points (FD 101). At early time ($t = 0.6$) particles are distributed heavily over the left region (maximum at $L = 0.6$) up to the final time of simulation, particles are mostly distributed on the right segment (maximum at $L = 1.8$). The numerical FD 101 solution is accurate for any segment except the sharp transition region. If we increase the number of discretization point it will increase the accuracy of the solution of the whole region (including the peak region). However, it requires more of computational effort due to the increase of ordinary differential equations (ODEs) needed to be solved.

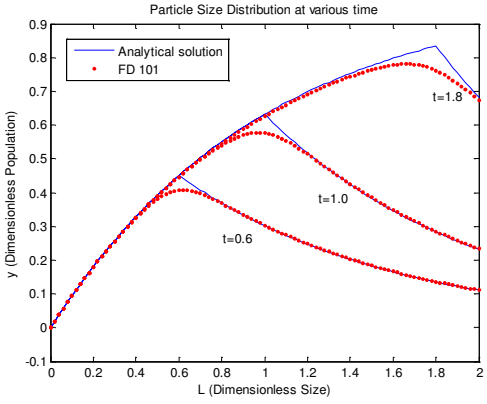

Fig. 2. PSD at various times using FD method

Other methods that can be used to solve this problem are, as previously mentioned Orthogonal Collocation (OC) and Orthogonal Collocation with Finite Element (OCFE). Detailed description of these method can be found in many literature such as Davis (1984), and Finlayson (1980).

Comparisons between the numerical solutions using OC, OCFE and FD are presented in Table 1 and Figure 3. Table 1 shows the comparative error results for the utilized methods, which are SE (Sum of Errors), AE (Average of Errors) and ME (Maximum of Errors), respectively.

In terms of computation time, even though the FD 101 consists of more ODEs than others, it gives reasonable computation time, only 1.37 s. On the other hand, the OCFE 16, which has only 16 ODEs contributes 2.21 s in computation. We can conclude that all methods described have reasonable computation time. The only problem is in the accuracy of the solution in the sharp transition region.

OCFE 31 gives the overpredicted result at the maximum point, whilst the other methods give underpredicted result at that point. OC method cannot be used because it only represents 8 points of solution and it does not cover the entire region proportionally. On the other hand, all OCFE methods perform better than the 101 points of FD method in the sharp transition region. It is indicated by the maximum of errors (ME) results of the OCFE 16 and the OCFE 31 which are less than the FD 101. Even though both methods use less number of collocation points, they successfully present accurate solution and especially in the peak region. It is revealed that the OCFE methods are superior compared to others.

Table 1 Comparative simulation results for case I

Method	Time (s)	SE	AE	ME
FD 101	1.37	8.21e-005	0.0091	0.00200
OC 8	0.90	0.0151	0.1230	0.07060
OCFE 7	0.19	3.3965e-004	0.0184	0.00210
OCFE 16	2.21	1.3513e-004	0.0116	0.00064
OCFE 31	3.34	1.0248e-004	0.0101	0.00110

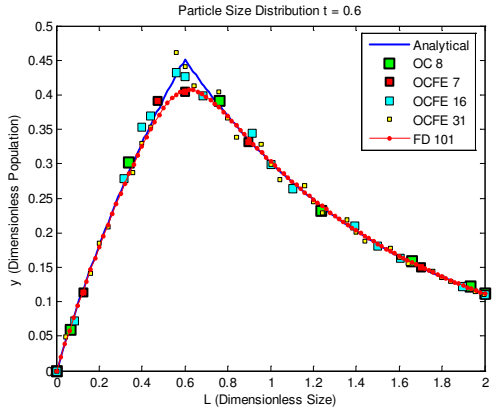

Fig. 3. PSD case I using various methods

At this point we can conclude that for the sharp transition region, the OCFE methods can be used instead of ordinary FD methods with reasonable

computational time and accurate solution. Another question that arose from this study is whether those methods are able to track a very dramatic changing profile as shown in the next case.

4.2 Case II: Size-independent growth only

Figure 4 shows that there is a very steep gradient profile in the particle size distribution. The particles are distributed mostly in region of $L = 1.7 - 2.3$. According to Liu et.al (2000) OCFE method can avoid spurious (unstable) responses under steady state conditions, however, it may fail for the transient model. In our case-II's simulation, unfortunately the OCFE method cannot be applied, since it gives unstable solution. On the other hand, the FD methods even with a very large point of discretization (401 and 801 points) represent inaccurate solutions in terms of the maximum value and also the particle distribution itself. As we can see from the Figure 5, there is a shift phenomena in particle distribution, and it shifts 0.5 unit of dimensionless size compared to analytical solution.

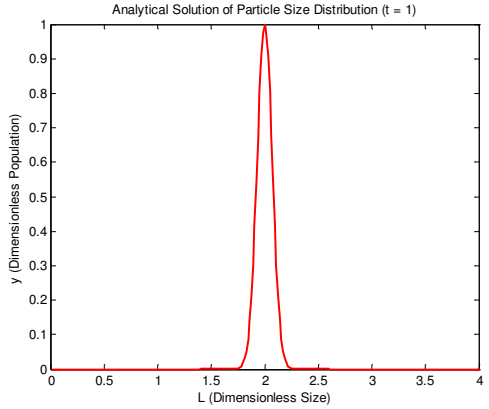

Fig. 4. Analytical solution for case II

Table 2 Comparative simulation results for case II

Method	Time (s)	SE	AE	Max(n)
FD 401	1.42	0.05334	0.2309	0.70717
FD 801	4.62	0.06844	0.2384	0.81654
FD 1201	11.07	0.05842	0.2417	0.86605
Wave 8	0.92	9.91e-6	0.0032	1.00

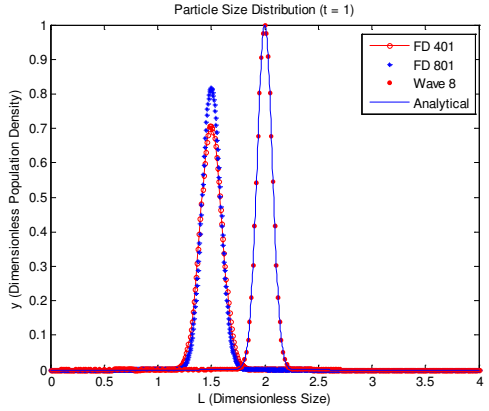

Fig.5. PSD case II using FD and Wavelet method

Other method must be considered here. Wavelet orthogonal collocation method is employed since it can represent a sharp transition region due to its good localization properties both on time and frequency. By using 8-level of wavelet approximation series (resolution), the solution can represent the accurate value at the peak point. Regarding the error parameters, wavelet solution appeared to be superior compared to previous methods. By using 8-level of resolution, we utilize 257 (256+1) wavelet collocation points for the solution and 255 numbers of differential equations. Given that the properties of wavelet which is capable of representing high localization both in space and frequency, allow the preview of the behaviour of the solution at certain time, from localization properties of the solution at previous time-step. Moreover, the computation time needed for FD methods is greater than the wavelet method.

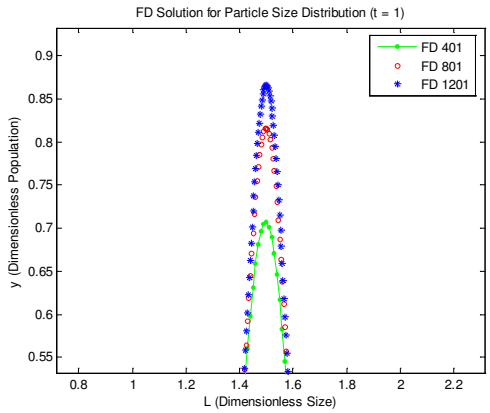

Fig. 6. Comparison of FD solution in case II

In our computational study, we used Runge-Kutta method in MATLAB® to utilize numerical integration of set of ODEs and all the parameters are set to their default value.

The exponential term in the initial condition contributes to the nature of the solution. The numerical solutions become highly non linear. At the early time ($t = 0$), the numerical solution agrees with the analytical solution. As the time increase the shifted solution becomes larger. We can conclude that the use of the FD technique for this case will generate highly inaccurate results. Furthermore we can see that the solution of the FD method still cannot track the dramatic change in particle size distribution even if there was no shift phenomena. As we can observe from Figure 6, the maximum value of $n(L,t) = 0.7$ for FD 401 and $n(L,t) = 0.8$ for FD 801. By increasing the number of discretization points from 801 to 1201 points, the solution still fails to move from 0.5 unit delay however it increased the maximum value of n from 0.816 to 0.866. As can be seen from the Figure 6, there was a small increase in reaching the maximum value by adding 400 points from 801 to 1201 points compared with 401 to 810 points. Comparison between Wavelet method and FD methods; and FD methods with OC and OCFE methods employed in this paper provide us some

insight into the superiority of wavelet in terms of accuracy and computation time.

Further research on wavelet application in chemical engineering field is essentially required. From the computational efficiency result shown, with the wavelet algorithms, the model is suitable to be employed in online control system. Model of population balance with multidimensional properties is necessary for certain cases. Even though from control engineers' perspective, low-order models are needed. The challenges for modelling a population balance with wavelet-based method is to define the suitable complexity for various cases and reduce the appropriate models for design the control strategies. Efforts will be made in future work to validate the results of this study using experimental data from the literature.

5. CONCLUSION

In this work, accurate, fast and general approach by wavelet method is the most efficient way to simulate the case of a very sharp transition phenomenon in population balance system. For case-I, the nucleation and size-independent growth only, the sharp change region can be described effectively by OCFE methods. It, however fail to simulate case-II where there was a very steep gradient in PSD profiles. The FD methods in both cases fail to provide accurate solutions. In case-II where there was a shift of 0.5-unit size they give incorrect prediction of the PSD profiles due to the presence of the highly non-linear term. Wavelet solution gives the fast, stable and accurate solution. The selection of the level of resolution being used depends on the characteristic of the solution itself. If the solution is highly non-linear, the level of resolution must be increased.

REFERENCES

Bertoluzza, S. and G. Naldi (1996). "A Wavelet Collocation Method for the Numerical Solution of Partial Differential Equations." Applied and Computational Harmonic Analysis **3**(1): 1-9.

Braatz, R. D. (2002). "Advanced control of crystallization processes." Annual Reviews in Control **26**(1): 87-99.

Chen, M.-Q., C. Hwang and Y-P. Shih. (1996). "A wavelet-Galerkin method for solving population balance equations." Computers & Chemical Engineering **20**(2): 131-145.

Daubechies, I. (1988). "Orthonormal bases compactly supported wavelets." Communications Pure and Applied Mathematics(41): 909-996.

Davis, M. E. (1984). Numerical Methods and Modeling for Chemical Engineers. New York, John Wiley and Sons.

Finlayson, B. A. (1980). Nonlinear Analysis in Chemical Engineering. New York, McGraw Hill.

Fujiwara, M., Z. K. Nagy, J.W. Chew and R.D. Braatz. (2005). "First-principles and direct design approaches for the control of pharmaceutical crystallization." Journal of Process Control **15**(5): 493-504.

Gupta, S. K. (1995). Numerical Methods for Engineers. New Delhi, New Age International.

Hangos, K. and I. T. Cameron (2001). Process Modelling and Model Analysis. London, Academic Press.

Hulburt, H. M. and S. Katz (1964). "Some problems in particle technology : A statistical mechanical formulation." Chemical Engineering Science **19**(8): 555-574.

Kostoglou, M. and A. J. Karabelas (1994). "Evaluation of Zero Order Methods for Simulating Particle Coagulation." Journal of Colloid and Interface Science **163**(2): 420-431.

Liu, Y., I. T. Cameron and F.Y. Wang. (2000). "The wavelet-collocation method for transient problems with steep gradients." Chemical Engineering Science **55**(9): 1729-1734.

Liu, Y. and I. T. Cameron (2001). "A new wavelet-based method for the solution of the population balance equation." Chemical Engineering Science **56**(18): 5283-5294.

Liu, Y. and I. T. Cameron (2003). "A new wavelet-based adaptive method for solving population balance equations." Powder Technology **130**(1-3): 181-188.

Liu, Y. and M. O. Tade (2004). "New wavelet-based adaptive method for the breakage equation." Powder Technology **139**(1): 61-68.

Motard, R. L. and B. Joseph (1994). Wavelet applications in chemical engineering. London, Kluwer Academic Publisher.

Nielsen, O. M. (1998). Wavelets in Scientific Computing. Department of Mathematical Modelling. Lyngby, Technical University of Denmark**:** 244.

Ramkrishna, D. (2000). Population balance : theory and applications to particulate systems in engineering. London, Academic Press.

Randolph, A. D. and M. A. Larson (1988). Theory of Particulate Processes : Analysis and Techniques of Continuous Crystallization. San Diego, Acadmic Press.

Rohani, S., M. Haeri and H.C. Wood. (1999). "Modeling and control of a continuous crystallization process Part 1. Linear and non-linear modeling." Computers & Chemical Engineering **23**(3): 263-277.

Shekunov, B. Y. and P. York (2000). "Crystallization processes in pharmaceutical technology and drug delivery design." Journal of Crystal Growth 211(1-4): 122-136.

Vanni, M. (2000). "Approximate Population Balance Equations for Aggregation-Breakage Processes." Journal of Colloid and Interface Science **221**(2): 143-16