Numerical Studies of Wavelet-based Method as an Alternative Solution for Population Balance Problems in a Batch Crystalliser

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Abstract: This paper deals with the numerical simulation studies of the nucleation and crystal growth process in a batch crystalliser. The population density functions may extend over orders of several magnitudes and the size distribution can be very sharp, thus accurate numerical solution of the population density functions can be challenging. The main interest for finding a more representative population balance solutions has motivated many researchers to develop many methods for the last four decades. In this paper, four methods have been implemented including wavelet-based method. The results obtained in three case studies, have demonstrated the wavelet scheme to be an alternative in providing accurate, fast and robust solutions.

Keywords: Crystallisation, Numerical scheme, Population Balance, Wavelet method

1. INTRODUCTION

Hulburt and Katz (1964) introduced a statistical mechanical formulation, known as population balance, for modelling the crystallisation process. The population balance equation (PBE) can be defined as a mathematical description characterising particles undergoing the mechanisms of birth, growth, death and leaving a certain particle phase space. In crystallisation, those mechanisms can be categorized as nucleation, growth, agglomeration and breakage. The population density functions may extend over orders of several magnitudes and the size distribution can be very sharp, thus the accurate numerical simulation of the population density functions can be challenging and has motivated several researchers in this area for decades to develop specialised algorithms for solving PBE, for example Ramkrishna (1981), Hounslow et al.(1988), Litster et al. (1995), Kumar and Ramkrishna (1996), Nicmanis and Hounslow (1996), and Mahoney and Ramkrishna (2002). All of these methods can be classified into four categories, such as method of weight residuals/orthogonal collocation, finite elements methods, finite difference schemes/discrete population balances methods, and other methods. There are major drawbacks from those methods such as computationally expensive, lack of stability and accuracy of the solution and in applicability of the solved models to be implemented in control based models. Extensive comparative discussion of those methods can be found in the literature (Kostoglou et al. 1994; Ramkrishna 2000; Vanni 2000). Therefore, the need for accurate, fast, robust and low order solution is essential for design, control and optimisation purposes.

This paper reports the application of wavelet based methods as an alternative, for solving population balance problems in a batch crystalliser condition. Previous study (Utomo et al. 2006) has been extended by comparing with finite difference based methods, such as upwind finite difference (U-FD), biased upwind finite difference (BU-FD), and method of weighted residuals, such as, orthogonal collocation with finite element method (OCFE). Different types of population balance are illustrated in the three cases discussed in this paper. They are having a high non-linearity, a steep-front profile, and stiff characteristic. The solutions are benchmarked with respects to their size (spatial grid points used), accuracy (mean and average error) and the computation time (t-CPU).

2. NUMERICAL SCHEMES

2.1 Previous Methods

Finite difference (FD) 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 was used to approximate the first partial derivative of population density over its size and converts the PDE into a set of ODEs.

In this paper, the upwind finite difference and biased upwind finite difference schemes were applied to effectively handle the instability and to avoid the spurious oscillation as generated by a centred FD scheme. The five-point (fourth-order accuracy) upwind and biased upwind on uniform grids were implemented as described in (Wouwer et al. 2005). Orthogonal collocation technique was developed 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 use of orthogonal polynomials is to reduce the error as the polynomial order increases (Gupta 1995; Hangos et al. 2001). OCFE is the combination method 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 elements. Selection of the elements size is therefore essential.

2.2 Wavelet-based methods

In 1992, Daubechies in her famous text-book, "Ten lectures on wavelets" (Daubechies 1992), predicted that a wavelet based software package to solve partial differential equations will be available in the market. The prediction has not been met as today there is no software available except for the Wavelet Toolbox in MATLAB®, which cannot be used for solving any partial differential equation (PDE).

The earliest wavelet application in chemical engineering was Wavelet Galerkin (WG). It was due to the work of Chen et al. (1996). Wavelet method with Galerkin scheme was utilised to solve the breakage population balance in a batch crystalliser. One of the challenges of WG is the expansion coefficients in WG was not specified in the physical space, while most of the PDEs can be directly solved in the physical space rather than converting and transforming its solution back to physical space. The second method was Wavelet Optimised Finite Difference (WOFD), developed by Jameson (1998). To date, this method has not been employed in the chemical engineering field. The third method was Wavelet Orthogonal Collocation (WOC). Its first application in chemical engineering was due to Liu and Cameron (2001). This method was successfully applied to solve the population balance and steep front concentration profiles in adsorption. However, WOC has not been applied for solving the complex cases which may involve the non-linearity and full dimensional variables. To sum up, the comparison of the three methods discussed are given below, which may initiate further development of a new wavelet-based numerical scheme for solving PDEs.

Table 1. Comparative components of three wavelet-based methods, WG: Wavelet Galerkin, WOFD: Wavelet Optimised Finite Difference and WOC: Wavelet Orthogonal Collocation

Comparative components	***WG***	***WOFD***	***WOC***
Basis calculation	Wavelet	**Physical**	**Physical**
BC treatment	Difficult	**Easy**	Moderate
Non-linearity handling	Difficult	**Moderate**	**Moderate**
Adaptive scheme	No	**Yes**	**Yes**
Computation capacity	Fixed	Fixed	**Reduced**

2.3 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 solution can be calculated from the collocation points. 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 gives 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:

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

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 et al. 1994). Detailed information about Daubechies orthonormal wavelets can be found in Daubechies (1988).

2.4 Wavelet Orthogonal Collocation(WOC)

This method was introduced 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 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:

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

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

3. CASE STUDIES

Three case studies of population balances, which have sharp transition phenomena in their particle size distribution in the batch crystallizer, were tested in this paper. Even though the case studies considered here are simple, the analytical solutions are available for comparison purposes.

3.1 Case I: Population balance with nucleation and size-independent growth

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

$$\frac{\partial n(x,t)}{\partial t} + G\frac{\partial n(x,t)}{\partial x} = B_0 \qquad G = 1;\ B_0 = \exp(-x) \tag{8}$$

where n is the number of particle (population density), x is the particle size, G is the growth rate, and B_0 is the nucleation rate.

With initial and boundary conditions such as:

$$n(t,0) = 0;\quad n(0,x) = 0 \tag{9}$$

The analytical solution for this case is:

$$n(t,x) = 1 - \exp(-x) \qquad x - t < 0$$
$$n(t,x) = \exp(-x)\left[\exp(-t) - 1\right] \qquad x - t > 0 \tag{10}$$

The dimensionless particle size L is defined as follow:

$$L = x/x,\max = [0:1]$$

Where: $x = [x,\min : x,\max] = [0:2]$

3.2 Case II: Population balance with size-independent growth only and initially seeded

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

$$\frac{\partial n(x,t)}{\partial t} + G\frac{\partial n(x,t)}{\partial x} = 0 \qquad G = 1 \tag{11}$$

With initial and boundary conditions such as:

$$n(t,0) = 0;\quad n(0,x) = \exp(-100(x-1)^2 \times 1/6.6) \tag{12}$$

The analytical solution for the second case is :

$$n(t,x) = \exp(-100(x - G.t - 1)^2 \times 1/6.6) \tag{13}$$

The dimensionless particle size L is defined as follow:

$$L = x/x,\max = [0:1]$$

Where: $x = [x,\min : x,\max] = [0:5]$

3.3 Case III: Seeded batch crystalliser with nucleation and growth

$$\frac{\partial n(L,t)}{\partial t} + G\frac{\partial n(L,t)}{\partial L} = B \tag{14a}$$

$$B = k_b\, M_T^{\,j}\, \Delta c^{\,b} \tag{14b}$$
$$G = k_g\, \Delta c^{\,g} \tag{14c}$$

where G is the growth rate, and B is the nucleation rate, k_g and k_b are the growth and nucleation constant, M_T is the suspension density, Δc is the supersaturation, and superscript g and b are the exponential constants for growth and nucleation rate respectively.

With initial and boundary conditions such as:

$$n(0,L) = n_0\,\delta(L - L_0)$$
$$n(t,0) = B/G \tag{15}$$

The supersaturation balance can be written as

$$\frac{d\Delta c}{dt} = \frac{3W_{S0}\,L_S^2\,G}{L_{S0}^3\,S} + \frac{3\rho\,k_v\,A_N\,G}{k_a} \tag{16}$$

where W_{S0} is the mass of seed crystals, $L_{S0}$is average size of seed crystals, L_S is the average size of growing crystals, S is the amount of solvent used, ρ is the crystal density, k_v is the volume shape factor, A_N is the total surface area of crystals, and k_a is the area shape factor.

The particle size L is defined as follow:

$$L = [L,\min : L,\max] = [400:1400]\,\mu m$$

4. RESULTS AND DISCUSSION

All the simulation results presented have been executed on an Intel(R) Core(TM) 2 CPU, with 2.00 GHz and 2.00 Gigabytes of RAM. A MATLAB® version 7.4.0.287 (R2007a) was used as the computation software to simulate the models. The built-in integrator of "ode15s" was utilised for integrating set of ordinary differential equations. The relative and absolute error of the integrator was specified at value of 10^{-3}.

4.1 Case I

The first case describes a simple population balance system which presents sharp front size distribution profiles. The PBE has a nucleation as function of size and a constant growth rate

and the analytical solution was available from (Chang et al. 1984). Four numerical schemes were applied in this case, their performance were tested in order to see the suitability, accuracy and stability of tested methods in handling the non-linearity and the sharp profile characteristic. The orthogonal collocation with finite elements (OCFE) scheme with 2 elements, which comprise of 31 grid points was applied. The other two methods were based on finite difference scheme. The upwind finite difference (U-FD) of 2 points and the biased upwind finite difference (BU-FD) of 5 points were employed. Both of them were discretised in space to generate 129 grid points. Lastly, wavelet orthogonal collocation (WOC) was used, and their performance were benchmarked with the analytical solution.

The average error (AE) was defined as the square root of sum of square error divided by the number of grid points. As it was calculated on an individual time basis, AE does not depend on the number of equations (grid points). The value of AE can illustrate the total absolute error of the grids for a certain time. A small value of AE may also illustrate a stable solution. While AE is the global error figure, the maximum error (ME) could show a local error or an overshoot from the reference value. ME was defined as the maximum value of the square root of sum of square error at a certain time.

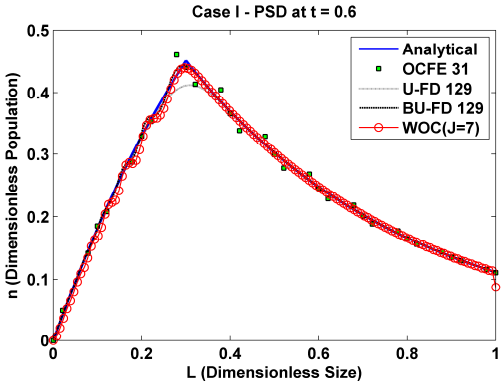

Fig. 1. Particle size distribution, case I at 0.6 seconds, simulated by four methods and compared with analytical solution.

Figure 1 shows a comparative particle size distribution (PSD) at 0.6 s. The particle distribution was initially zero and the nucleation start to generate nuclei and at the same, the born nuclei grow at a constant rate of 1.0. OCFE 31 profile produced a slightly overestimated particle density than the analytical solution. While, all the other numerical schemes, including WOC, with the same resolution gave comparable results. The simplest two points upwind scheme gave an under predicted population at the peak point, while the simulation results from biased upwind and wavelet are equivalent in term of minimising the error at the peak point. From the AE point of view, U-FD was the least while BU-FD was the first accurate then followed by WOC. When the wavelet level (J) was increased from 7 to 9, the ME values were about the same but an increased accuracy was achieved by 62%, consequently at the same time the computation time (t-CPU) was 440% higher.

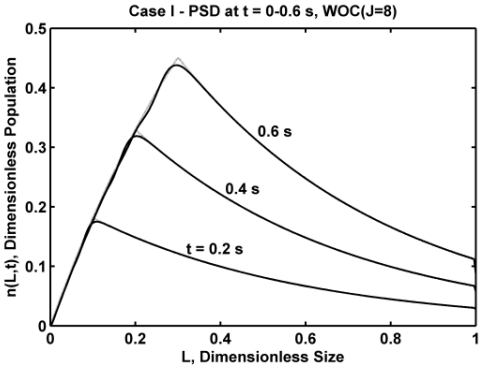

Fig. 2. Particle size distribution of WOC (J=8) at 0.2, 0.4 and 0.6 second, the black line: WOC solution and grey line: analytical solution.

In this simulation, all the computation time was short (less than 5 seconds) because all the methods have the same structure of a matrix form. Matrix to matrix calculation was superior than loop calculation (using *for* loop) for its computation time and its adaptability to a more complex case. Figure 2 shows the solutions produced by WOC (J=8) at various time from t = 0.2, 0.4 and 0.6 seconds. At this point, it can be concluded that OCFE methods can be with reasonable accuracy level and WOC method can be employed as an equivalent alternative solution for handling the case of sharp fronts profile caused by non-linear nucleation function. A question that arose is whether these methods are able to track a very sharp profile as shown in the next case.

Table 2. Numerical performance results for case I, N: grid points, AE: average error, ME: maximum error, t-CPU: computation time

Case I @ 0.6 s	N	AE	ME	t-CPU
OCFE (2)	31	0.4383	0.7861	< 1s
U-FD (2)	129	0.0082	0.0391	< 1s
BU-FD (5)	129	0.0031	0.0120	< 1s
WOC (J=7)	129	0.0064	0.0243	< 1s
WOC (J=8)	257	0.0033	0.0242	1.4 s
WOC (J=9)	513	0.0024	0.0239	4.4 s

4.2 Case II

In a seeded batch crystalliser, where the nucleation can be minimised, particle size distribution will be controlled only by the initial condition of seeding and a crystal growth. In this case, the crystal growth was assumed again constant, and the seed condition was artificially made to present a very sharp front of particle size distribution. The previous study done by Utomo, et al. (2006) reported that OCFE method cannot be applied as the unstable solutions were obtained. Moreover, the upwind finite difference scheme gave delayed solutions. Therefore, only WOC method as an equivalent method was tested in this case. The effect of wavelet resolution (J) and vanishing moments (M) were observed to closely study its performance.

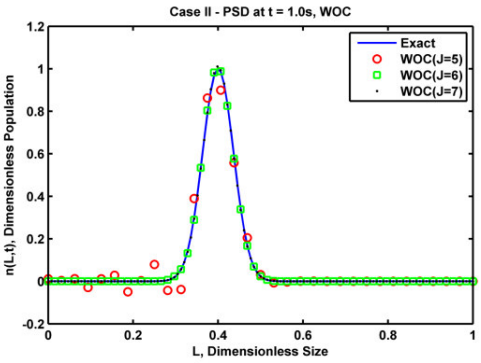

Fig. 3. Particle size distribution of WOC at t = 1s, using various J =5,6,7.

Figure 3 shows the WOC solutions with various wavelet resolution (J=5, 6, and 7) as compared to the exact analytical solution. It is clear that except for J=5, WOC presented a good validation result with high accuracy. WOC for J=5, however, not only gave a high ME but also oscillation and a negative value problem. The final solution of J=5 was stable, but early oscillation recorded the highest average error in the solution. The same problem was mentioned by (Muhr et al. 1996), can be easily rectified by decreasing the spacing or utilising adaptive gridding.

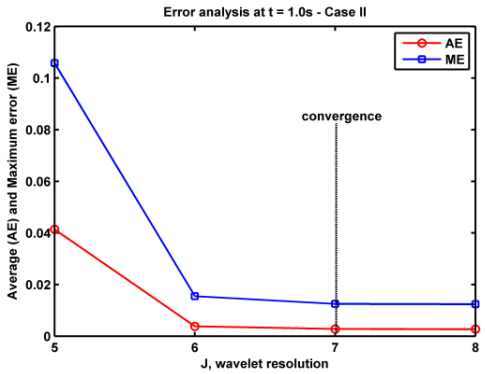

Fig. 4. The error analysis at t = 1s for various J, where AE: average error and ME: maximum error.

The convergence issue is important for numerical simulation. The WOC method for the tested resolution could easily give good convergence at a resolution as low as 6. Both AE and ME could be used for error analysis to show the convergence at a certain time. As shown in Figure 4, the convergence was actually reached as the J was increased from 6 to 7 and further increased in resolution would not give any improvement in the accuracy. The effect of vanishing moments (M) could only be observed, when the lowest resolution was applied. M = 0 and 1 were the most optimal values in giving the lowest level of error. At this stage, it can be summarised that WOC could have been also employed as an alternative for a very sharp front's profile. The selection of resolution is more sensitive to the computational performance rather than the sequential choice of vanishing moments. Selection of M becomes sensitive only when the symptom of instability was observed. To demonstrate WOC capabilities as an alternative method, the more complex population balance in a batch crystalliser case study was performed in the next section.

Table 3. Numerical performance results for case II; M: vanishing moments, N: grid points, AE: average error, ME: maximum error, t-CPU: computation time

Case II @ 1.0 s	M	N	AE	ME	t-CPU
WOC (J=5)	0	33	0.0369	0.0933	< 1.0 s
	1		0.0368	0.0933	
	2		0.0406	0.0931	
	3		0.0414	0.1058	
WOC (J=6)	0-3	65	0.0038	0.0155	< 1.0 s
WOC (J=7)	0-3	129	0.0028	0.0125	< 1.0 s
WOC (J=8)	0-3	257	0.0027	0.0124	1.3 s

4.3 Case III

The last case presents a seeded batch crystalliser with capacity of 25.5 kg solvent, running in an isothermal condition for batch time of 6000 seconds. The nucleation and growth kinetics are described in (14b) and (14c). The initial condition was the seed condition at the average size of 500 μm and the boundary condition is outlined in (15). To solve the system, a population balance equation (14a) coupled with the mass (supersaturation) balance for the solute and solid phase as in (16), thus the dynamic of crystal size distribution (CSD) can be computed. All the parameters used in this case were adopted from (Tavare et al. 1986).

For illustrative purposes, Figure 5, shows the profile of supersaturation and crystal growth rate during 6000 seconds batch operation. The initial condition of 0.015 kg/kg solvent would give a corresponding crystal growth of 1.68×10^{-10} m/s. The WOC method with J = 7 and M = 1 was employed and a reasonable result was obtained in Figure 6 as the experimental results was not available.

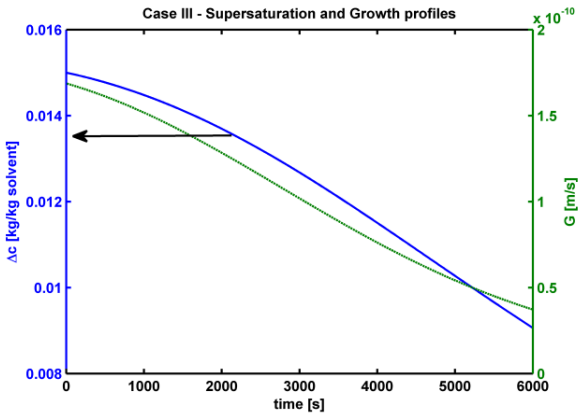

Fig. 5. Supersaturation (left) and crystal growth rate (right) profiles for case III up to 6000 seconds.

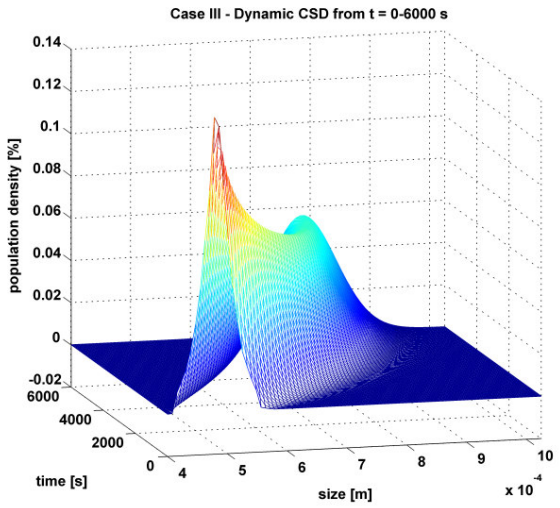

Fig. 6. Dynamic crystal size distribution for case III, for 400-1000 μm in size and 0-6000 seconds.

5. CONCLUSIONS

In this paper, three case studies which present a very sharp size distribution profile have demonstrated the potential of wavelet-based numerical scheme as an alternative in providing accurate, fast and robust solutions. Further research on a new wavelet numerical scheme and wavelet application in chemical engineering field is essentially required and promising. From the computational efficiency result shown, with the WOC algorithm, the model is suitable to be employed in online control system, however, from control engineers' perspective, low-order models are needed.

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