
Towards the Optimal Reconstruction of a Distribution from its Moments

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Summary. The optimal reconstruction of a distribution knowing only a finite number of its moments is an extremely important and still unsolved question for many fields of science (chemical and process engineering, electronic engineering, nuclear physics, image processing, biotechnology, . . .). Several methods have been proposed and corresponding mathematical formulations have been introduced in the literature during the last decades. Nevertheless, all these are generally limited to particular, simple cases and require specific assumptions. It is indeed extremely difficult from a theoretical point of view (all moments are in theory required for a correct reconstruction) as well as from a practical point of view (ill-posed inverse problem) to find an accurate and relatively fast method which can be applied to all scientific areas. In the present paper, different possible methods (prescribed functions, discrete method, spline-based reconstruction) allowing such a reconstruction are explained, compared in terms of efficiency and accuracy, and finally validated using a distribution corresponding to the model of an ideally-mixed batch crystallizer.

1 Introduction and state of the art

In the last 10 years, a renewed interest can be observed in the scientific community dealing with chemical engineering applications concerning moment-based determination of Particle-Size Distributions (PSD). This is in particular due to the fact that, when external features like turbulent flow properties play an important role for the process under investigation, fast and numerically efficient methods must be employed to describe the population interacting with this flow. Acceptable computational costs are typical for standard moment methods and for related approaches like the quadrature method of moments (QMOM) and its direct alternative (DQMOM). For all such methods, only a finite number of moments associated with the real distribution are finally determined by the numerical procedure. Therefore, after having computed these moments, it is necessary to reconstruct in the best possible way the full, real distribution corresponding to the resulting PSD. Since the PSD generally constitute the key result to judge the quality of the process, the high importance of the reconstruction procedure appears clearly.

This inverse problem is well-known and has been often considered during the last hundred years, not only in the field of chemical engineering. Different solutions have been proposed and tested. Nevertheless, no really satisfactory method can be found in the literature up to now. For most researchers in chemical engineering dealing with fundamental aspects, this is due to the fact that, mathematically, all the moments up to infinity are requested in order to obtain a perfect reconstruction. As such, the problem considered in this work would not have any practicable solution. At the other end, for chemical engineers dealing with very applied questions, one possible solution concerning the reconstruction, often used in practice, is to assume a priori the shape of the distribution (Gaussian, log-normal, beta function...) and to find a best fit of the small number of parameters needed to fully determine this assumed distribution.

Building on top of this experience, methods are developed in this work which should be easily usable in practice to obtain the best possible PSD, but without imposing a priori the shape of the distribution. In this way, complex PSD can be computed naturally and at acceptable numerical costs using the described procedure. This is made possible in particular by revisiting the mathematical conditions underlying this inverse reconstruction problem, while explicitly taking into account the supplementary information concerning the constraints underlying any real PSD found in chemical engineering. By combining those

with optimal methods of applied mathematics and algorithms, new possibilities appear for moment-based reconstruction. In the paper, the available literature on this subject is first considered, going back to Smoluchowski [1] up to present time [2, 3], to show that both approaches (mathematical view: “this is impossible”; practical view: “just use an assumed shape”) offer more possibilities than what has been used up to now. In the mathematical literature, the finite-moment problem has been mainly studied analytically, proving in particular the existence of solutions for certain classes of functions (see for example [4, 5]). However, a thorough treatment of this problem from the point of view of Numerical Mathematics is still missing and efficient algorithms are not available.

2 Approaches for reconstructing a PSD

Examples of results obtained using a priori shapes for the PSD will be first presented in order to illustrate the advantages (fast and easy computations) and drawbacks (limitation to simple shapes, a priori knowledge needed about the solution) of this approach.

Secondly, a discrete method based on a time-dependent update of the distribution together with or after the computation of the moments will be presented. This method is very fast, numerically efficient and easy to implement, but cannot always be extended to all possible physical processes encountered in chemical engineering. In this case, by solving numerically the moment equations, the trajectories of the system are determined and can be used to recover the time-dependent birth and nucleation profiles during the process. Knowing the initial particle distribution, a simple numerical algorithm is implemented to add new particles and shift appropriately the existing ones.

Finally, a highly flexible solution based on spline functions is presented to solve the reconstruction problem. For this, a spline-based reconstruction is introduced to describe the distribution, without any a priori hypothesis concerning its shape. Using the information contained in the moments, a very good approximation of the real distribution can be obtained at an acceptable computational cost. The practical numerical procedure underlying all these approaches will be presented in detail.

3 Parameter fitting using a-priori function shapes

In this section, examples of results obtained using a-priori shapes for the PSD are presented in order to illustrate the advantages (fast and easy computations) and drawbacks (limitation to simple shapes, a-priori knowledge needed about the solution) of this approach.

Indeed, the fastest and the easiest method to reconstruct a PSD knowing only its moments is fitting it to a prescribed mathematical function (such as Gaussian, log-normal, β -function... [6, 7]) by using a certain number of low-order moments obtained through the population balance model, for example the standard method of moments (SMM), first proposed by Hulburt and Katz [8] and used since then by many authors [9, 10, 11, 12]. The SMM has been also employed in [3] and further extended to more general versions (QMOM in [13] and DQMOM in [14]), since the SMM has proven to be insufficiently accurate for a number of cases. Even if these newer methods are more general in nature, the reconstruction of the PSD must be carried out exactly in the same manner as using SMM. Reconstruction based on known functions is in that case a quite powerful and fast method, allowing quite often to determine single-peak PSDs with an excellent accuracy. A PSD with several peaks can be reconstructed using a linear superposition of single-peak reconstructed PSDs, as demonstrated afterwards.

Due to the fact that the size distribution of a particulate product is an essential criterion for assessing the properties and quality of that product [7], the assumed shape of the PSD curve must of course accurately fit the corresponding data. The resulting accuracy can be measured quantitatively by comparing either to a direct numerical solution of the Particle Balance Equations (PBE) or to experimental results. To illustrate this procedure, such a comparison is presented in Fig. 1 for a two-peaked PSD, comparing the direct solution of the PBE (reference solution) with the reconstruction based on known functions, either Gaussian or β -shaped [15]. Here, the two peaks correspond respectively to nucleated crystals (left part, smaller size) and to initial seed crystals after growth during a crystallization process (right part, larger size). In that case, the direct solution of the PBE directly delivers the two peaks, while our reconstruction procedure is carried out separately for each peak to increase the accuracy. In the left hand plot, two Gaussian-functions are used whereas β -functions are chosen in the right hand figure. As can be seen in these figures, the Gaussian function reproduces the shape of the nucleated crystals more exactly, showing that the nucleated crystals have a rather symmetric distribution. On the other hand, the crystals initially introduced as seeds at the beginning of the process are much better represented by the β -function.

This is not a surprise since the experimentally-measured PSD of the initial seed crystals follows already very nearly a β -function shape. Such β -functions are widely used in engineering applications, for example to model turbulent flows. The bottom picture in Fig. 1 illustrates the best possible combination of the reconstructed PSDs for this particular example. For all cases, only the first three moments are needed for the fitting procedure, so that the corresponding population models are easily solved for.

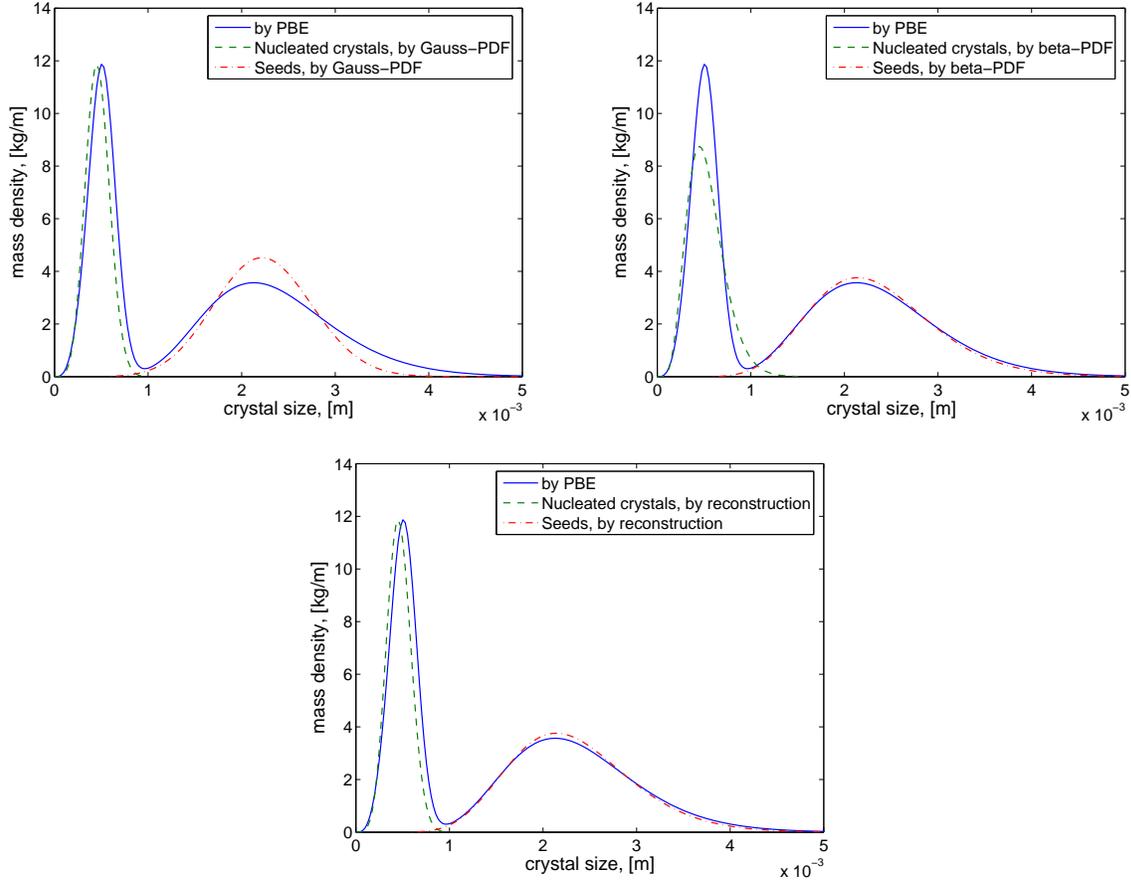


Fig. 1. Direct solution of the PBE, used as a reference (solid line) and moment-based reconstruction based on known functions. Top-left: reconstruction based solely on Gaussian functions. Top-right: reconstruction based solely on β -functions. Bottom: best possible combination for the reconstruction (Gaussian function for nucleated crystals and β -function for seeds after growth).

As a conclusion, the reconstruction of PSDs using a few low-order moments and considering a known function is a very efficient method, since the solution is obtained instantaneously. The obtained accuracy can be very high when using the right functional shape. But this method requires a lot of information concerning the expected distribution. Thus, this method is only accurate and should always be retained when the properties of the final PSD are quite well-known (for example when considering a small variation compared to a known process). On the other hand, when considering a completely new application with unknown PSD, this method cannot reasonably be retained. In that case, alternatives must be derived, as proposed afterwards.

4 Discrete methods

A discrete method based on a time-dependent update of the distribution together with or after the computation of the moments is now presented. For this purpose, a simplified dynamic model of an ideally-mixed batch crystallizer is first obtained by assuming an overall size-independent growth rate G and an overall nucleation rate B . Phenomena like attrition, breakage and agglomeration are neglected. Under these assumptions, the resulting population balance becomes [9]:

$$\frac{\partial F(t, x)}{\partial t} = -G \frac{\partial F(t, x)}{\partial x}, \quad (1)$$

where t is the time, x represents the characteristic particle size and $F(t, x)$ is the number density function. The following boundary and initial conditions apply:

$$F(t, 0) = \frac{B}{G}, \quad F(0, x) = F_{\text{seeds}}(x), \quad (2)$$

where $F_{\text{seeds}}(x)$ is the form of the initial seeds' distribution. The mass balance of the liquid phase is modeled by the integro-differential equation

$$\frac{dm_{\text{liq}}}{dt} = -3\rho_s k_v G \int_0^\infty x^2 F(t, x) dx. \quad (3)$$

The growth rate G is assumed to depend on the supersaturation only and it is described by the power-law

$$G = k_g (S - 1)^g. \quad (4)$$

Here, k_g is an overall crystal growth rate constant and S denotes the actual degree of supersaturation.

If crystals are already dispersed in the crystallizing medium, secondary nucleation can occur at supersaturation levels which are significantly lower than those at which primary nucleation takes place [7]. Its rate can be described by a power-law expression

$$B = k_b (S - 1)^b \int_0^\infty x^3 F(t, x) dx, \quad (5)$$

where k_b is generally assumed to be related to the stirring power.

The distributed model (1)-(5) can be greatly simplified by converting it into a moment model [16]. From (1) - (2), a set of ODEs for the moments of the PSD (here actually a crystal size distribution) can be derived. The moments are defined by

$$\mu_i(t) = \int_0^\infty x^i F(t, x) dx, \quad i = 0, 1, 2, \dots \quad (6)$$

The zeroth moment μ_0 corresponds to the overall number of crystals, and the third moment μ_3 is proportional to the volume of the crystalline material in the crystallizer. By partial integration, it follows from (1), (2) that

$$\frac{d\mu_0(t)}{dt} = B, \quad \frac{d\mu_i(t)}{dt} = iG\mu_{i-1}(t), \quad i = 1, 2, \dots \quad (7)$$

In this approach, only the first 4 moments are used since they can be calculated independently from the higher moments. The solution of the system (3) - (5), (7) is not expensive in terms of computational time, but it describes accurately only the dynamics of the overall characteristics of the PSD. The information concerning the internal property (in this case the crystal size x) is lost after applying transformation (6). However, the approximate form of the PSD can be recovered by a simple numerical algorithm, which exploits the fact that crystal nucleation takes place at negligible particle size, i.e. using Eq. (2). The numerical solution of the reduced system, Eqs. (3) - (5), (7), delivers information about the mass of the dissolved material m_{liq} at discrete time instances Δt during the batch process. By Eqs. (4) and (5) the corresponding growth and nucleation rate for each time instance can be calculated. Using the recovered growth and nucleation time trajectories, the time-evolution of the boundary condition, Eq. (2), can also be recovered. This is sufficient for an accurate reconstruction of the PSD.

The recovery procedure starts from the known size-discretized initial condition for the seeds $F_{\text{seeds}}(x)$. For each time step Δt particles at zero size are added, according to the boundary condition (Eq. 2), and the complete distribution is then shifted towards larger sizes, according to the growth rate profile. Fig. 2 illustrates schematically this iterative process in time.

5 Using general functions for the reconstruction

In this section, a highly flexible solution based on general functions is presented to solve the reconstruction problem. For this, a spline-based reconstruction is introduced to describe the distribution, without any a priori hypothesis concerning its shape. Using the information contained in the moments, a very good approximation of the real distribution can be obtained at an acceptable computational cost. The practical numerical procedure is now presented in detail. Let $[a, b] \subset \mathbb{R}$ be divided into n subintervals $[x_i, x_{i+1}]$, $i = 1, \dots, n$ with $a = x_1 < x_2 < \dots < x_n < x_{n+1} = b$. A spline $s^{(k)}(x) : [a, b] \rightarrow \mathbb{R}$ of degree k is a function with the following properties:

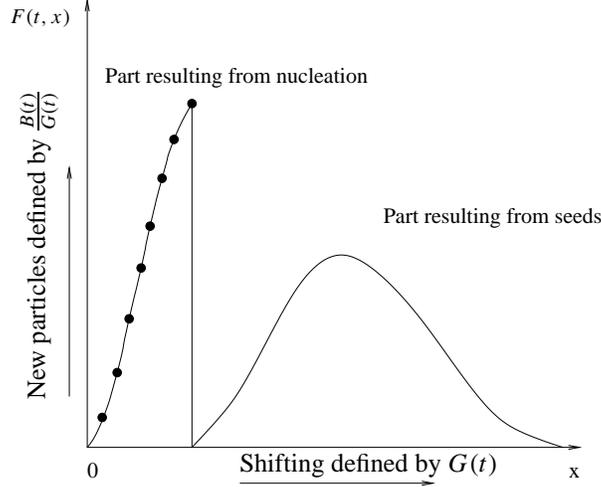


Fig. 2. PSD recovery using the discrete method.

- $s^{(k)}(x)$ is in each subinterval $[x_i, x_{i+1}]$, $i = 1, \dots, n$ a polynomial of degree k ,
- $s^{(k)}(x)$ is $(k - 1)$ -times continuously differentiable in $[x_1, x_{n+1}]$.

Very popular in numerical integration are cubic splines, i.e. $k = 3$. We will apply them here for the reconstruction of PSDs. Thus, the ansatz is

$$s_i(x) = \sum_{j=0}^3 s_{ij}(x - x_i)^j, \quad x \in [x_i, x_{i+1}], \quad i = 1, \dots, n, \quad (8)$$

i.e., $4n$ unknown coefficients s_{ij} are to be determined. Using cubic splines assumes that the PSD to recover is smooth in (a, b) . Otherwise, lower-order splines should be applied at non-smooth points of the PSD. This supplementary problem is the subject of present work.

The smoothness of the spline at the nodes x_i , $i = 2, \dots, n$ gives three conditions for each node leading altogether to $3(n - 1)$ equations. Three conditions have to be posed at each boundary. Thus, $4n - (3(n - 1)) - 6 = n - 3 =: m$ conditions have to be described by given moments of the PSD. The other way around, if m moments are available, the PSD can be approximated by a spline with $m + 3$ subintervals. At the end, one obtains a $4n \times 4n$ linear system of equations to determine the unknown coefficients in (8).

For choosing the boundary conditions, one can use the fact that the PSD $f(x)$ has a compact support $\text{supp}(f)$. Let

$$\text{supp}(f) \subset [a, b], \quad (9)$$

then one has the conditions $s_1(x_1) = s_n(x_{n+1}) = 0$. If no other information is available, one should use in the first and the last subinterval a linear spline by setting

$$s_{12} = 0, s_{13} = 0, s_{n2} = 0, s_{n3} = 0. \quad (10)$$

If it is known that the PSD has a smooth transition to zero, one can specify instead of (10)

$$s'_1(x_1) = 0, s''_1(x_1) = 0, s'_n(x_{n+1}) = 0, s''_n(x_{n+1}) = 0. \quad (11)$$

An a priori knowledge of an optimal interval $[a, b]$ might not be available in applications. On the one hand, it is crucial to choose the interval $[a, b]$ such that (9) is fulfilled. On the other hand, for the quality of the reconstruction, it is advantageous if $[a, b]$ is chosen as small as possible, thereby respecting (9). Our algorithm adjusts the interval $[a, b]$ dynamically to reach this goal. E.g., if it is found that the reconstructed function is smaller than a prescribed tolerance in the last subinterval $[x_n, x_{n+1}]$, then the end point b is moved to the left by setting $b := (x_n + x_{n+1})/2$. After this redefinition of $[a, b]$, the nodes in the new interval $[a, b]$ are redistributed equi-distantly. Then, the spline reconstruction is computed in the new interval. The adjustment of the interval might take place several times until a suitable final interval $[a, b]$ is found.

The linear system of equations for the moment reconstruction is ill-conditioned. Instead of using the inverse of the system matrix to compute the solution, we use the pseudo inverse where small singular values are neglected. This is a well known approach for regularization of ill-conditioned problems. This

regularization smoothes the oscillations of the reconstructed PSD and thus prevents large negative values. Since only a finite number of moments are available and there are in general data errors, one cannot expect that the ill-posed finite moment problem will lead to a perfectly non-negative reconstruction of the PSD. Instead, we allow very small negative values. The smallest singular values are neglected iteratively until this criterion is fulfilled. If in addition no reduction of the current interval $[a, b]$ is recommended, the iterative process is stopped and the final reconstruction is obtained.

6 A numerical study with the different methods

To finally illustrate the accuracy of all these approaches (advantages, problems and limitations), we consider as a test-case a given, three-peaked PSD as a first but already quite complex problem (see Fig. 3). This PSD was computed by solving directly a Population Balance Equation for crystallization. The size of the crystals is in $[0, 5 \cdot 10^{-3}]$ m and the maximal value of the PSD is around $2.8 \cdot 10^7$ 1/m. The first eight moments are computed from the PSD obtained directly using the PBE with a discretization based on numerical quadrature. These moments μ_i are given with a unit m^i .

$$\begin{aligned} \mu_0 &= 5.5911475010957845 \cdot 10^4 & \mu_1 &= 9.7447189797086963 \cdot 10^1 \\ \mu_2 &= 2.2713126695535327 \cdot 10^{-1} & \mu_3 &= 6.0281801524505118 \cdot 10^{-4} \\ \mu_4 &= 1.7044920031329059 \cdot 10^{-6} & \mu_5 &= 5.0010225203552623 \cdot 10^{-9} \\ \mu_6 &= 1.5051001861147765 \cdot 10^{-11} & \mu_7 &= 4.6186266332179558 \cdot 10^{-14}. \end{aligned}$$

Parameter fitting using a-priori function shapes. The result obtained with 3 β -functions is presented in Fig. 3. Using a prescribed function shape, it is possible to reconstruct instantaneously the most important features of the PSD. However, the use of a β -function is not the perfect choice for the leftmost peak. There, the difference between the PSD and its reconstruction is rather large (more than 20% difference on the peak value). For the present example, it would be easily possible to find a better approximation by using different a priori shape functions for the left peak. However, this solution is often impossible in practice since the PSD is not known and there is no possibility to compare different reconstructions with the exact PSD.

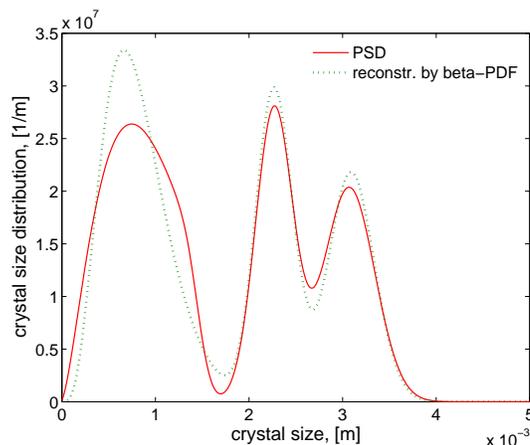


Fig. 3. Reconstruction by parameter fitting using a-priori function shapes. Thick solid line: PSD, used to determine the moments and giving the reference solution. Dotted line: reconstruction using 3 β -functions based on 3×3 moments.

Discrete method. On the left part of Fig. 4, the employed, known distribution of the initial seeds is presented. The thick line on the right figure presents the numerical solution of the PBE, when the spatial variable is discretized using backward differences. This scheme of discretization is stable, but also introduces a non-negligible amount of numerical diffusion. This can be clearly seen if the seeds' distributions at the beginning and at the end of the process are compared - their peaks should have the same height, but the final distribution should be shifted towards a certain larger size. Here, the distribution at the end of the process is shifted, but at the same time it is more dispersed. The dotted line on the right figure shows the PSD reconstructed by the discrete method. Only the temporal evolution

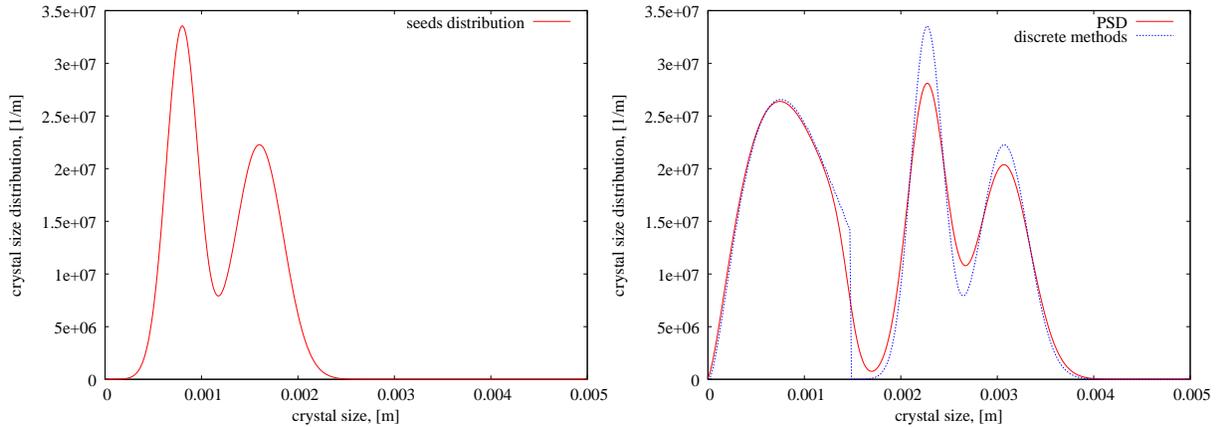


Fig. 4. Reconstruction by the discrete method. Thick solid line, left figure: form of the seeds' distribution. Thick solid line, right figure: numerical solution of the PSD using a backward difference discretization of the PBE. Dotted line, right figure: reconstruction with the discrete method using 4 moments.

of the first four moments has been used. The peaks are higher, which shows that this method does not introduce a significant numerical diffusion.

Altogether, the presented discrete method from Sec. 4 is successfully applied for a relatively simple, but largely used model which describes a batch-wise particulate process. The obtained results not only show a good agreement with the numerical solution of the distributed model, but they also preserve the initial form of the seeds crystals. The largest part of the disagreement obtained on Fig.4 (right) is due to the numerical diffusion introduced when solving numerically the PBE. Improvements concerning this point are presently being implemented.

Spline-based reconstruction. The results obtained with the spline reconstruction are presented in Fig. 5. Since $m = 8$ moments are available, one can use a spline with $n = 11$ subintervals. On the left boundary, the particle density distribution does not have a smooth transition to zero, i.e., the first and second right-sided derivatives at $x = 0$ are not zero. Hence, the first interval is reconstructed by a linear spline, see (10). On the right hand side, the PSD possesses a smooth transition to zero such that the boundary conditions (11) are applied. Since the algorithm should work without exact a priori knowledge of the support of the particle density function, it is started with the large interval $[0, 10^{-2}]$ m.

Figure 5 shows that the algorithm detects that initially in the right part of the interval, the reconstructed function is almost zero and it successively decreases the length of the interval down to around $[0, 4.2 \cdot 10^{-3}]$ m. The final reconstruction is obtained after 20 iterations and it reproduces quite well the most important features of the reference PSD.

7 Conclusions and perspectives

The reconstruction by parameter fitting using a priori prescribed shape functions is currently the most popular approach because of its simplicity. This will not change in the near future. However, this approach possesses some drawbacks which have been described in Sec. 3.

The interest of the discrete method for simple configurations has been demonstrated on the considered example. In that case, the reconstruction is obtained almost without any additional computational cost. This same discrete method, Sec. 4, has been also applied recently for a more complicated process model, where part of the existing crystals, which are below a certain critical size, are dissolved in a fines trap and the clear liquid is fed back to the crystallizer. In this case, the method had to be applied in each step of the numerical integration of the reduced system in order to obtain the numerical flux of the dissolved particles. Consequently, the necessity of repetitively applying the discrete method partly eliminates the advantage of a reduced calculation time compared to the numerical solution of the distributed system. Further investigations have the goal to improve the performance of the discrete approach in such complex cases.

A challenge in the further development of the spline reconstruction described in Sec. 5 consists in the a posteriori detection of non-smooth points in the PSD and the automatic use of low order splines at such points.

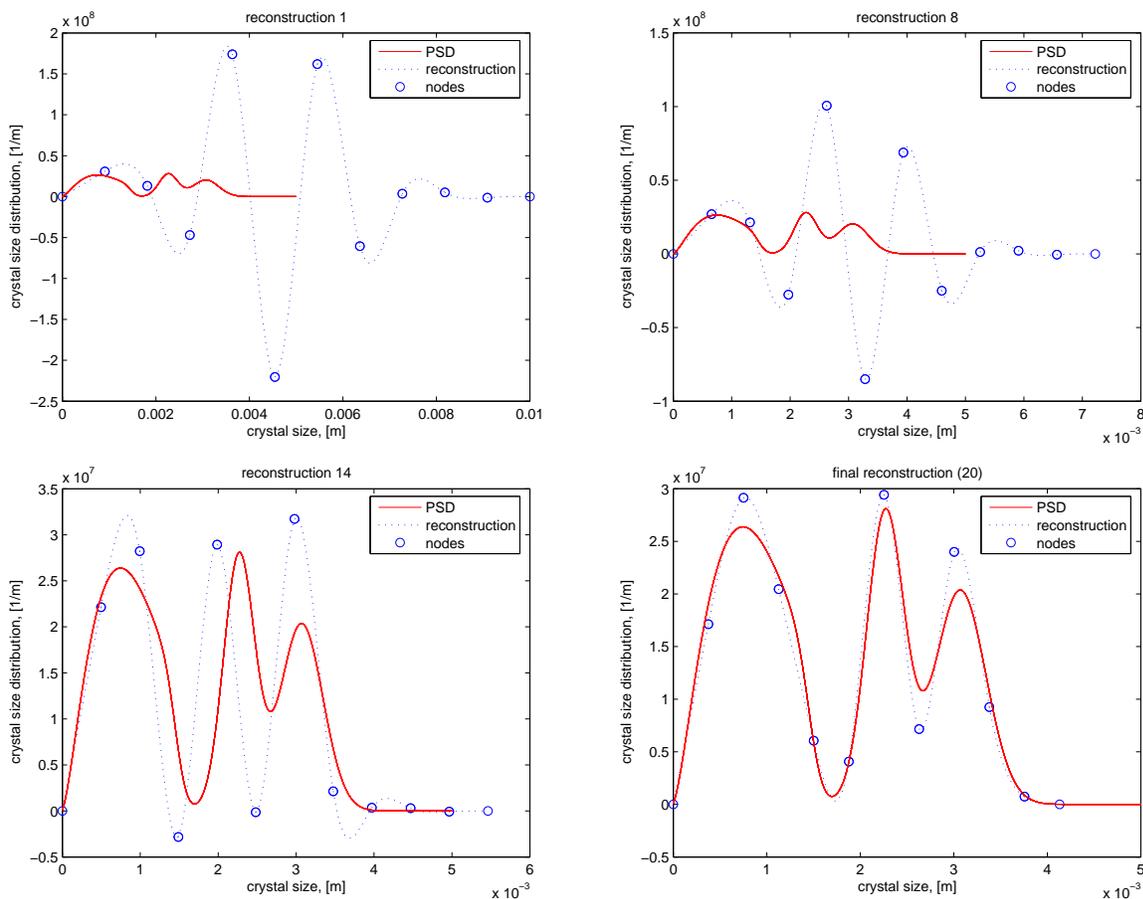


Fig. 5. Reconstruction by splines using eight moments. We show the current estimation after 1, 8, 14 and 20 (convergence) iterations.

In spite of all these limitations and remaining questions, different solutions leading to a fast and efficient reconstruction of a PSD knowing only a finite number of its moments have been proposed and demonstrated on an example in this work.

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