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Research Article

The Asymptotic Behavior of Particle Size Distribution Undergoing Brownian Coagulation Based on the Spline-Based Method and TEMOM Model

Qing He [1] and Mingliang Xie [2]

[1] *Key Laboratory of Distributed Energy Systems of Guangdong Province, School of Chemical Engineering and Energy Technology, Dongguan University of Technology, Dongguan 523808, China*
[2] *State Key Laboratory of Coal Combustion, School of Energy and Power Engineering, Huazhong University of Science and Technology, Wuhan 430074, China*

Correspondence should be addressed to Qing He; heqing@dgut.edu.cn

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In this paper, the particle size distribution is reconstructed using finite moments based on a converted spline-based method, in which the number of linear system of equations to be solved reduced from $4m \times 4m$ to $(m+3) \times (m+3)$ for $(m+1)$ nodes by using cubic spline compared to the original method. The results are verified by comparing with the reference firstly. Then coupling with the Taylor-series expansion moment method, the evolution of particle size distribution undergoing Brownian coagulation and its asymptotic behavior are investigated.

1. Introduction

Particle size distribution (PSD) is one of the most important properties of aerosol particles, including transport, sedimentation, and so on [1]. It is also of utmost interest in many industrial applications, such as powder preparation and particle synthesis [2, 3]. The evolution of the PSD undergoing different dynamic processes is usually described in the framework of population balance equations (PBEs) mathematically [4], which have a strong nonlinear structure in most cases and cannot be solved analytically. With a high-computational efficiency, the moment-based method has become a powerful tool for investigating aerosol microphysical processes, in which cases some statistical characteristics of the PSD, namely, the moments of the PSD, are obtained [5]. However, the detailed information about the target PSD is out of reach. Theoretically, the PSD is equal to the moments of all orders. The proof about the uniqueness of reconstruction in the case all moments are known is given with an appropriate condition that the range of the PSD is in a finite interval [6]. But in practice, only a finite number of moments are obtained.

Using a given number of moments to reconstruct the PSD is known as the finite-moment problem or inverse problem in mathematical analysis [7]. Generally, this problem is distinguished between the three types for the monovariate case: the Hausdorff moment problem with the PSD supported on the closed interval $[a, b]$, where $[a, b]$ are the lower and upper limits of the domain of PSD; the Stieltjes moment problem with the PSD supported on $[0, +\infty)$; and the Hamburger moment problem with the PSD supported on $(-\infty, +\infty)$ [8]. Until now, there exist several frequently used reconstruction methods in the literature mainly for the Hausdorff moment problem, including but not limited to parameter-fitting method, Kernel density function-based method, maximum entropy method, and spline-based method. The parameter-fitting method is to assume the PSD as a simple function (i.e., log-normal distribution or gamma distribution), where the parameters in the function are determined by the given low-order moments [7]. It is the fastest and easiest method but with drawbacks that need a priori knowledge about the solution and limited to simple shapes, even though a weighted sum of different simple functions can be used [9]. The kernel density function-based method is a positivity-preserving representation and can be regarded as the development of parameter-fitting method, which approximates the PSD by a superposition of weighted kernel density functions [10]. This method gives rise to an ill-posed problem for determining weights, and a large number of available moments are needed to ensure accuracy. Based on the maximization of the Shannon entropy or the minimization of the relative entropy from information theory, the maximum entropy method is a notable method which needs relatively less knowledge of the prior distribution or the number of moments compared to the previous two methods [8, 11]. With the advantage of no priori assumptions on the shape of the PSD as well as that the needed number of moments only depends on that of interpolation nodes, the spline-based method proposed by John et al. [7] has attracted some researchers' attention, such as the investigation on particle aggregation and droplet coalescence [12, 13]. And an adaptive spline-based algorithm with a wider application for nonsmooth and multimodal distributions was developed later [6]. More relevant research about the comparisons between these methods can be found in the literature [14, 15].

In this paper, we will use the spline-based method to reconstruct the PSD coupling with PBEs describing Brownian coagulation in the free molecule regime and continuum regime. Compared to the original method, the number of linear system of equations to be solved is significantly reduced through substituting the continuous conditions. The correctness of this new treatment is verified by comparing with the reference results in [7]. Then with the moments obtained by the Taylor-series expansion moment method (TEMOM) [16], the evolution of PSD due to Brownian coagulation and its asymptotic behavior are investigated.

2. Theory and Modeling

2.1. Modeling of Spline-Based Method. In the original method, the support of the target PSD $[a, b]$ is divided into m subintervals: $a = x_1 < x_2 < \cdots < x_{m+1} = b$. In each subinterval, the PSD is approximated by a spline (piecewise polynomial) $s_i^{(l)}(x)$ of degree l; thus, there exist $(l+1)m$ unknowns. For cubic spline ($l = 3$), the splines $s^{(l)}(x)$ and their first and second derivatives are continuous at each node x_i ($i = 2, 3, \ldots, m$), which give $3(m-1)$ conditions. With the smooth boundary conditions, which means $s^{(l)}(x)$ and their first and second derivatives are null at nodes x_1 and x_{m+1}, there still require $m-3$ additional conditions, which have to be supplemented by the known moments. Then, a $4m \times 4m$ ill-conditioned linear system is obtained. In order to improve the accuracy of calculation, the interval should be set as small as possible, which is controlled by tol_{red}. For example, the last (or the first) subinterval is divided into n smaller subintervals: $x_m = x_{m1} < x_{m2} < \cdots < x_{mn} = x_{m+1}$; if the ratio of 2-norm of $s^{(l)}(x_{mn})$ to the maximum of $s^{(l)}(x)$ is less than tol_{red}, the last node is reset as $x_{m+1} = (x_m + x_{m+1})/2$. Furthermore, tol_{neg} and tol_{sing} are introduced to guarantee that the value of $s^{(l)}(x)$ is nonnegative. More detailed procedure is shown in [7].

In this paper, we use a converted ansatz for $s^{(l)}(x)$ to reduce the number of the linear system. For cubic spline, the second derivative in each node is set as L_i, then $s''(x)$ can be written in the following form using linear interpolation:

$$s_i''(x) = L_i \frac{x - x_{i+1}}{x_i - x_{i+1}} + L_{i+1} \frac{x - x_i}{x_{i+1} - x_i}, \quad x \in [x_i, x_{i+1}], \ (i = 1, 2, \ldots, m). \tag{1}$$

Then, $s(x)$ and their first derivatives can be gotten through integrating:

$$\begin{aligned} s_i'(x) &= \frac{L_i}{2} \frac{(x - x_{i+1})^2}{x_i - x_{i+1}} + \frac{L_{i+1}}{2} \frac{(x - x_i)^2}{x_{i+1} - x_i} + C_{i1}, \\ s_i(x) &= \frac{L_i}{6} \frac{(x - x_{i+1})^3}{x_i - x_{i+1}} + \frac{L_{i+1}}{6} \frac{(x - x_i)^3}{x_{i+1} - x_i} + C_{i1} x + C_{i2}, \end{aligned} \tag{2}$$

where C_{i1} and C_{i2} are integral constants. With the continuity of the spline and their first derivatives at x_i ($i = 2, 3, \ldots, m$), we can get

$$\begin{aligned} C_{i1} &= C_{11} + \sum_{j=2}^{i} L_j \frac{(\Delta x_{j-1} + \Delta x_j)}{2}, \\ C_{i2} &= C_{12} - \sum_{j=2}^{i} L_j \frac{(\Delta x_{j-1} + \Delta x_j)}{2} \frac{(x_{j-1} + x_j + x_{j+1})}{3}, \end{aligned} \tag{3}$$

in which Δx_i is the length of the ith subinterval and C_{11} and C_{12} are related to the left boundary conditions. Thus, the sum of the number of moments and boundary conditions needed to solve the equations is $m + 3$.

Usually, we consider that the value of PSD out of the support $[a, b]$ is small enough and can be set as zero:

$$\begin{aligned} s_1(x_1) &= 0, \\ s_m(x_{m+1}) &= 0, \end{aligned} \tag{4}$$

and the first derivatives are denoted as

$$\begin{aligned} s_1'(x_1) &= q_1, \\ s_m'(x_{m+1}) &= q_2, \end{aligned} \tag{5}$$

where q_1 and q_2 are zero for smooth boundary conditions. Then, (3) can be simplified by substituting the left boundary conditions:

$$\begin{aligned} C_{i1} &= q_1 + \sum_{j=1}^{j=i} L_j d_j, \\ C_{i2} &= -q_1 x_1 - \sum_{j=1}^{j=i} L_j d_j e_j. \end{aligned} \tag{6}$$

Together with the right boundary conditions, we can get the following formula:

$$\begin{aligned} q_1 - q_2 + \sum_{i=1}^{i=m+1} L_i d_i &= 0, \\ q_1 x_1 - q_2 x_{m+1} + \sum_{i=1}^{i=m+1} L_i d_i e_i &= 0, \end{aligned} \tag{7}$$

in which d_i and e_i are given as follows ($i = 2, 3, \ldots, m$):

$$\begin{aligned} d_i &= \frac{\Delta x_{i-1} + \Delta x_i}{2}, \\ d_1 &= \frac{\Delta x_1}{2}, \\ d_{m+1} &= \frac{\Delta x_m}{2}, \\ e_i &= \frac{x_{i-1} + x_i + x_{i+1}}{3}, \\ e_1 &= \frac{2x_1 + x_2}{3}, \\ e_{m+1} &= \frac{x_m + 2x_{m+1}}{3}. \end{aligned} \quad (8)$$

The kth order moment M_k of the PSD is defined as follows:

$$M_k = \int_0^\infty x^k f(x) dx. \quad (9)$$

Thus, the kth order moment of $s(x)$ is

$$\begin{aligned} &\int_{x_i}^{x_{i+1}} x^k \sum_{i=1}^{m} s_i(x) dx \\ &= \sum_{i=1}^{m} \left(\frac{-L_i}{6\Delta x_i} \left(I_{i4} - 3x_{i+1} I_{i3} + 3x_{i+1}^2 I_{i2} - x_{i+1}^3 I_{i1} \right) \right. \\ &\quad \left. + C_{i1} I_{i2} + \frac{L_{i+1}}{6\Delta x_i} \left(I_{i4} - 3x_i I_{i3} + 3x_i^2 I_{i2} - x_i^3 I_{i1} \right) + C_{i2} I_{i1} \right), \end{aligned} \quad (10)$$

in which I_i are

$$\begin{aligned} I_{i1} &= \frac{x_{i+1}^{k+1} - x_i^{k+1}}{k+1}, \\ I_{i2} &= \frac{x_{i+1}^{k+2} - x_i^{k+2}}{k+2}, \\ I_{i3} &= \frac{x_{i+1}^{k+3} - x_i^{k+3}}{k+3}, \\ I_{i4} &= \frac{x_{i+1}^{k+4} - x_i^{k+4}}{k+4}. \end{aligned} \quad (11)$$

In order to represent L_i explicitly, (10) is arranged as follows:

$$\begin{aligned} \int_{x_i}^{x_{i+1}} x^k \sum_{i=1}^{m} s_i(x) dx &= q_1 \frac{x_{m+1}^{k+2} - x_1^{k+2}}{k+2} - q_1 x_1 \frac{x_{m+1}^{k+1} - x_1^{k+1}}{k+1} \\ &\quad + \sum_{i=1}^{m} (G_i L_i + H_i L_{i+1}), \end{aligned} \quad (12)$$

where

$$\begin{aligned} G_i &= -\frac{I_{i4} - 3x_{i+1} I_{i3} + 3x_{i+1}^2 I_{i2} - x_{i+1}^3 I_{i1}}{6\Delta x_i} \\ &\quad + d_i \frac{x_{n+1}^{k+2} - x_i^{k+2}}{k+2} - d_i e_i \frac{x_{n+1}^{k+1} - x_i^{k+1}}{k+1}, \\ H_i &= \frac{I_{i4} - 3x_i I_{i3} + 3x_i^2 I_{i2} - x_i^3 I_{i1}}{6\Delta x_i}. \end{aligned} \quad (13)$$

Now together with (7), a $(m+3) \times (m+3)$ linear system for L_i, q_1, and q_2 is obtained. Next, we will discuss the number of moments that should be supplemented (note that, in this paper, all cases calculated with q_1 and q_2 are zero):

(1) If q_1 and q_2 are unknowns, $(m+1)$ moments are needed to solve these equations.

(2) If q_1 and q_2 are zero or any other constants given, $(m-1)$ moments are needed; if the value of $s''(x)$ at boundary is given (such as smooth conditions in [7], namely, $L_1 = L_{m+1} = 0$), $(m-3)$ moments are needed. And in this case, the order of the coefficient matrix is $(m+1) \times (m+1)$ or $(m-1) \times (m-1)$.

(3) If q_1 and q_2 obey some relationships, for example, $q_1 = (s_1(x_2) - s_1(x_1))/\Delta x_1$, $q_2 = (s_m(x_{m+1}) - s_m(x_m))/\Delta x_m$, then $L_1 = -L_2/2$ and $L_{m+1} = -L_m/2$ can be derived and $(m-1)$ moments are needed.

For quadratic spline, we can also get a $(m+1) \times (m+1)$ linear system in the same way by denoting that

$$\begin{aligned} s_i'(x) &= l_i \frac{x - x_{i+1}}{x_i - x_{i+1}} + l_{i+1} \frac{x - x_i}{x_{i+1} - x_i}, \\ x &\in [xi, \ xi+1], \quad (i = 1, 2, \ldots, m), \end{aligned} \quad (14)$$

where l_i is the first derivative in each node. The corresponding $s(x)$ and linear system are as follows:

$$\begin{aligned} s_i(x) &= \frac{l_i}{2} \frac{(x - x_{i+1})^2}{x_i - x_{i+1}} + \frac{l_{i+1}}{2} \frac{(x - x_i)^2}{x_{i+1} - x_i} + c_{i1}, \\ &\sum_{j=1}^{j=m+1} l_j d_j = 0, \\ &\int_{x_i}^{x_{i+1}} x^k \sum_{i=1}^{m} s_i(x) dx = \sum_{i=1}^{m} (g_i l_i + h_i l_{i+1}), \end{aligned} \quad (15)$$

where

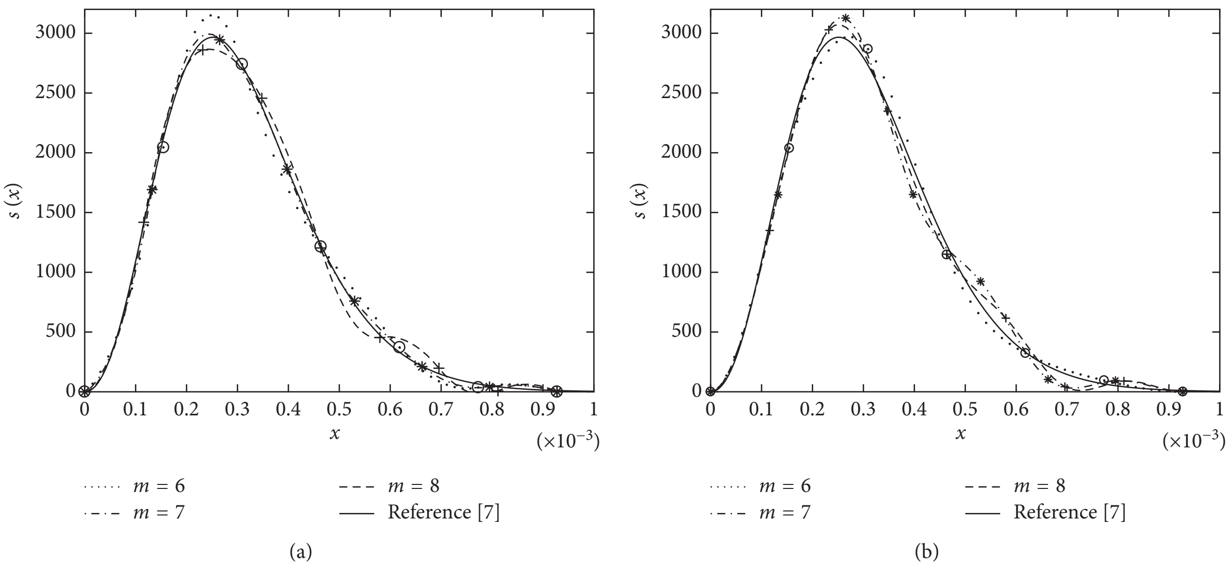


Figure 1: Reconstruction of the PSD for different moments (m) about Example 2.1 in [7] by using (a) quadrature spline and (b) cubic spline.

$$g_i = -\frac{I_{i3} - 2x_{i+1}I_{i2} + x_{i+1}^2 I_{i1}}{2\Delta x_i} + d_i \frac{x_{n+1}^{k+1} - x_i^{k+1}}{k+1},$$
$$h_i = \frac{I_{i3} - 2x_i I_{i2} + x_i^2 I_{i1}}{2\Delta x_i}, \quad (16)$$
$$c_{i1} = \sum_{j=1}^{j=i} l_j d_j.$$

2.2. Modeling of PBE and TEMOM. The population balance equation describing irreversible Brownian coagulation with continuous monovariable can be written as follows [17]:

$$\frac{\partial n(v,t)}{\partial t} = \frac{1}{2}\int_0^v \beta(v, v - v_1) n(v_1, t) n(v - v_1, t) dv_1 - \int_0^\infty \beta(v_1, v) n(v, t) n(v_1, t) dv_1, \quad (17)$$

where $n(v,t)$ is the number density function of the particles with volume from v to $v + \mathrm{d}v$ at time t and $\beta(v_1, v)$ is the collision frequency function between particles with volume v and v_1. In the free molecule and continuum regime, $\beta(v, v_1)$ are represented separately as

$$\beta_{\mathrm{FM}}(v, v_1) = B_1 \left(\frac{1}{v} + \frac{1}{v_1}\right)^{1/2} \left(v^{1/3} + v_1^{1/3}\right)^2,$$
$$\beta_{\mathrm{CR}}(v, v_1) = B_2 \left(\frac{1}{v^{1/3}} + \frac{1}{v_1^{1/3}}\right)\left(v^{1/3} + v_1^{1/3}\right), \quad (18)$$

in which $B_1 = (3/4\pi)^{1/6}(6k_{\mathrm{b}}T/\rho_{\mathrm{p}})^{1/2}$ and $B_2 = 2k_b T/3\mu$, where k_{b} is the Boltzmann constant, T is the temperature, ρ_{p} is the particle density, and μ is the gas viscosity.

With the definition of the kth order moment, M_k, (17) is transformed to a series of original differential equations by multiplying both sides with v^k and then integrating over all particle sizes:

$$\frac{dM_k}{dt} = \frac{1}{2}\int_0^\infty \int_0^\infty \left[(v + v_1)^k - v^k - v_1^k\right] \times \beta(v, v_1) n(v, t) n(v_1, t) dv dv_1. \quad (19)$$

Using the Taylor-series expansion technology to approximate the collision frequency function and fractional moments, the moment equations are closed without any other artificial assumption [16, 18]. In the original TEMOM model, the first three moments can be obtained easily using the fourth-order Runge-Kutta method with M_1 remaining constant due to the mass conservation requirement. The corresponding higher and fractional moments are as follows [19]:

$$M_k = \frac{M_1^k}{M_0^{k-1}}\left[1 + \frac{k(k-1)(M_C - 1)}{2}\right], \quad (20)$$

where $M_C = M_0 M_2 / M_1^2$ is a dimensionless moment. Obviously, the reconstruction depends heavily on the reliability of known moments. Based on the log-normal size distribution assumption, the maximum relative error for M_k of this model is discussed by Xie [19], and the results demonstrate that the error of M_k for $k \le 2$ with a small standard deviation is acceptable. Furthermore, theoretical analysis of the PBE is feasible because of the relative simple form of this model [20, 21], and the explicit asymptotic solutions are as follows:

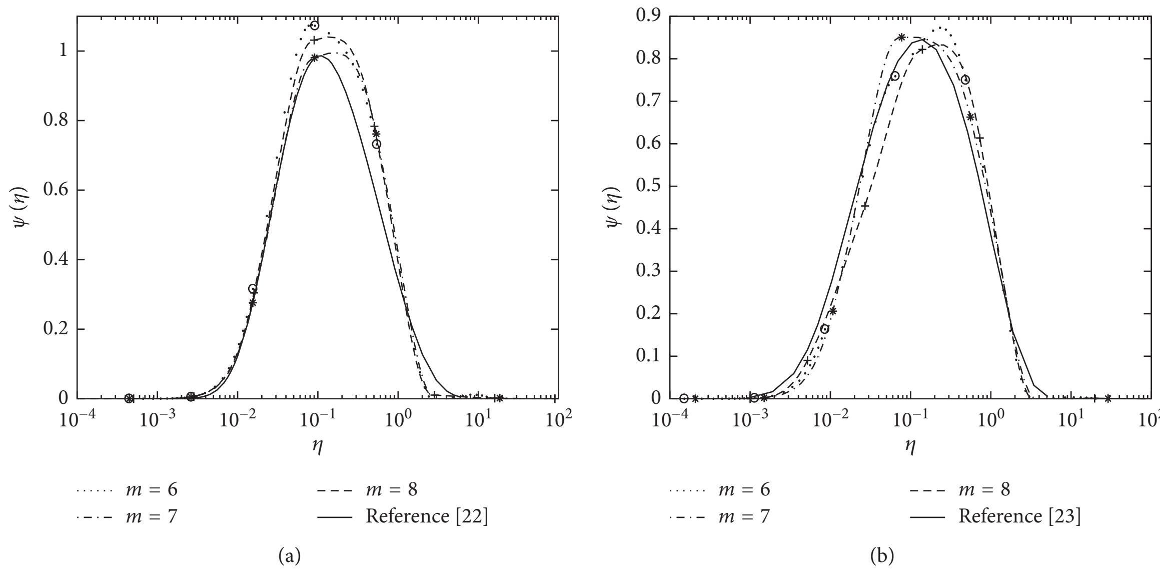


Figure 2: Reconstruction of $\psi(\eta)$ for different moments (m) by using cubic spline in the (a) free molecule regime and (b) continuum regime.

$$M_0|_{FM} \longrightarrow 0.313309932 \times B_1^{-6/5} M_1^{-1/5} t^{-6/5},$$
$$M_2|_{FM} \longrightarrow 7.022205880 \times B_1^{6/5} M_1^{11/5} t^{6/5},$$
$$M_0|_{CR} \longrightarrow \frac{81}{169} B_2^{-1} t^{-1},$$
$$M_2|_{CR} \longrightarrow \frac{338}{81} B_2 M_1^2 t, \tag{21}$$

and M_C tends to a constant 2.200126847 or 2, respectively. Using the similarity transformation $\eta = v/(M_1/M_0)$, the PSD can be arranged as follows:

$$n(v,t) = \frac{M_0^2}{M_1} \psi(\eta). \tag{22}$$

According to the theory of self-preserving, $\psi(\eta)$ does not change with time at a large t [1], and its moments only depend on k and M_C:

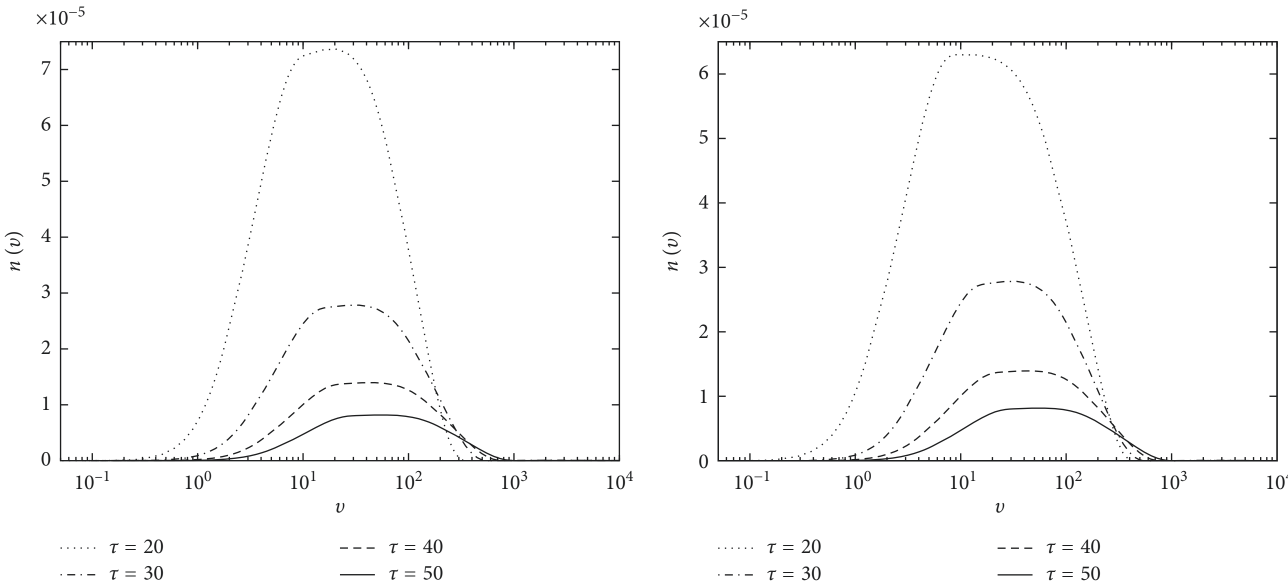
$$m_k = \int_0^\infty \psi(\eta)\eta^k d\eta = 1 + \frac{k(k-1)(M_C - 1)}{2}. \tag{23}$$

Then, $\psi(\eta)$ can be approximated by $s(\eta)$ using the spline-based method, and the asymptotic behavior of $n(v, t)$ is also known together with (21) and (22).

3. Results and Discussions

One difficulty of the inverse problem is the ill-conditioned coefficient matrix of the linear system. Another is that the value of $s(x)$ is nonnegative. By using the pseudoinverse routine, a least-squares solution of the linear system is obtained, in which the singular values smaller than tol_{sing} are set as zero (see Remark 4.2 in John et al. [7]). Moreover, the parameter α is introduced to avoid large difference in the order of magnitude. In this paper, we will follow this treatment. Figure 1 shows the results of the reconstruction about Example 2.1 in [7] by using quadrature spline and cubic spline proposed in this paper. And the parameters tol_{red}, tol_{neg}, and tol_{sing} are set as the same of those in the literature to maintain consistency. It should be noted that the tolerance values have an influence on the results [7, 14]. The great agreements with the references verify the validity of this new converted method. However, an underlying flaw is that only the continuity of $s(x)$ is necessary in practice. Moreover, the sensitivity of tol_{sing} to solution may increase when the number of the linear system sharply decreases. It can also be seen that some inflexion points appear with m increasing. This may be caused by the increasing condition number of the linear system.

The reconstruction of $\psi(\eta)$ in the free molecule regime and continuum regime for different moments (m) by cubic spline is shown in Figure 2, where the references are from Lai et al. [22] and Friedlander and Wang [23]. The initial interval is set as $[1e-5, 10]$, and the spacing of adjacent nodes are equidistant logarithmically. Both the left and right boundaries are adjusted according to the comparison results of $||s(\eta_{mn})||^2/\max(s(\eta))$ and tol_{red}, with $\text{tol}_{\text{red}} = 1e-2$ for $m = 6$ and $1e-4$ for $m = 7$ or 8, respectively. Besides, $\text{tol}_{\text{red}} = -1e-2$ and the initial value of tol_{sing} are set as $1e-36$. In addition to the first three integral moments m_0, m_1, and m_2, the fractional moments $m_{1/3}$, $m_{2/3}$, $m_{4/3}$, and $m_{5/3}$ are chosen to reconstruct $\psi(\eta)$, for the reason that these fractional moments with volume-based variable are proportional to the integral moments with length-based variable. It can be seen that the results for $m = 7$ show relatively small differences compared to the references. Generally, the differences may be caused by two parts: one is the error of TEMOM model and the other is the error of spline-based method. Scaling the moments M_k and time t by $M_k{}^* = M_k M_{00}^{(k-1)}/M_{10}^k$, $\tau = tB_1 M_{00}^{5/6} M_{10}^{1/6}$, or $\tau = tB_2 M_{00}$, the evolution of dimensionless $n(v, t)$ for $m = 7$ at long time is presented in Figure 3 with the initial conditions given as



(a) (b)

FIGURE 3: The evolution of dimensionless $n(v, t)$ with time for $m = 7$ in the (a) free molecule regime and (b) continuum regime.

$M^*_{00} = 1$, $M^*_{10} = 1$, and $M^*_{20} = 4/3$. Obviously, the particle number decreases and the average volume increases with time advancing due to coagulation.

4. Conclusion

By establishing the ansatz $s(x)$ on the basis of the continuity of second derivation, the number of linear ill-conditioned system can be reduced significantly from $4m \times 4m$ to $(m+3) \times (m+3)$ for $(m+1)$ nodes by using cubic spline, although only the continuity of $s(x)$ is necessary in practice. Then, coupling with the asymptotic solutions of TEMOM [20] and the theory of self-preserving, the evolution of the PSD due to Brownian coagulation in the free molecule regime and continuum regime and its asymptotic behavior are obtained easily.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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