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# Analytical Approach For Solving Population Balances: A Homotopy Perturbation Method

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## Abstract

In the present work, a new approach is proposed for finding the analytical solution of population balances for aggregation and fragmentation process. This approach is relying on idea of Homotopy Perturbation Method (HPM). The HPM solves both linear and nonlinear initial and boundary value problems without nonphysical restrictive assumptions such as linearization and discretization. It gives the solution in the form of series with easily computable solution components. The outcome of this study reveals that the proposed method can avoid numerical stability problems which often characterize in general numerical techniques related to this area. Several examples including Austin's kernel available in literature are examined to demonstrate the accuracy and applicability of the proposed method. In addition to it, the analytical solution to two new kernels [power-law kernel in fragmentation and the Ruckenstein/Pulvermacher kernel in aggregation] are also introduced.

**Keyword:** Particles; Population Balance Equation; Homotopy Perturbation Method; Analytical Solution.

## 1 Introduction

Aggregation and fragmentation represent two of the most basic particulate processes. Aggregation refers to the formation of a cluster from the combination of two smaller clusters; fragmentation refers to the reverse of this process. Together they represent two of the most elementary mechanisms that determine the size distribution of a particulate population. Many problems in the physical sciences and engineering involve either of both of these processes: pharmaceutical granulation, grinding of solids, polymerization/depolymerization, flocculation, and sol-gel processing are examples of such systems (Ramkrishna, 2000; Litster and Ennis, 2013; Ho et al., 2018; Ismail et al., 2018; Kaur et al., 2018a,b). In both breakup and aggregation the mathematical problem is an integral-differential equation, differential in time, integral in the size of the particles, whose solution by analytical techniques remains to this day a challenge. Breakage is described by two functions,  $a(n)$ , which gives the rate of fragmentation of particle size  $n$  (we take the particle mass as the size coordinate), and a function  $k(m|n)$  that gives the probability that parent size  $n$  produces a fragment of size  $m$ .

The breakage as well as aggregation models will be described by two functions. For defining the breakage model, the rate of breakup of the parent particle,  $a(m)$ , where  $m$  is the size of the particle, and the distribution of fragments,  $k(m|n)$  that gives the number of fragments of size  $m$  produced by a parent of size  $n$ . The breakage function  $k(m|n)$  obeys the following normalization conditions:

$$\int_0^n k(m|n)dm = \bar{f}, \quad (1.1)$$

$$\int_0^n mk(m|n)dm = n. \quad (1.2)$$

The first condition gives the average number of fragments,  $\bar{f}$ , and is the same for all parent sizes  $m$ ; the second condition expresses mass conservation between the parent particle and the fragments. A physically

realistic fragmentation model requires  $\bar{f} \geq 2$ . The governing equation for the size distribution of population that undergoes the above fragmentation process is given by the fragmentation equation,

$$\frac{\partial c(m, t)}{\partial t} = \underbrace{-a(m)c(m, t)}_{\text{death of particle of size } m} + \underbrace{\int_m^\infty a(n)k(m|n)c(n, t)dn}_{\text{birth of particle of size } m}, \quad (1.3)$$

where  $c(m, t)$  is the concentration of particles whose mass is in  $(m, m + dm)$  (we normalize the mass  $m$  by its mean value at time 0).

The governing equation for aggregation is

$$\begin{aligned} \frac{\partial c(t, m)}{\partial t} = & \underbrace{\frac{1}{2} \int_0^m g(m-n, n)c(t, m-n)c(t, n)dn}_{\text{birth of particle of size } m \text{ due to aggregation of particles of sizes } m-n \text{ and } n} \\ & - \underbrace{\int_0^\infty g(m, n)c(t, m)c(t, n)dn}_{\text{death of particles of sizes } m \text{ due to aggregation of particles of sizes } m \text{ and } n}, \quad t \in [0, T], \quad m, n \in (0, \infty) \quad (1.4) \end{aligned}$$

where  $\beta(m, n)$  is the aggregation kernel, which describes the rate at which the particles of sizes  $m$  and  $n$  coagulate to form a particle of size  $m + n$ . It is non-negative and symmetric function of its arguments.

The fragmentation and aggregation equations have both been the subject of several investigations and the analytical solutions have been obtained for a small number of special cases. Various solutions to the fragmentation equation have been given (Simha, 1941, 1956; Tobolsky, 1957; Ziff and McGrady, 1986; Ziff, 1991; Ernst and Szamel, 1993; Singh and Hassan, 1996). Few studies related to the numerical approximations such as Singh et al. (2014, 2015, 2016a,b, 2018a,b) have been also listed here. The study of the aggregation has an even longer history that begins with Smoluchowski's formulation of this equation and its solution for  $\beta(m, n) = 1$  (Smoluchowski, 1917). A small number of cases has been solved analytically and that include the constant ( $\beta = 1$ ), sum ( $\beta = m + n$ ) and product ( $\beta = mn$ ) kernels under monodisperse or other special initial conditions. A comprehensive review of these classical kernels was given by Leyvraz (2003). Other developments include moment generating functions Krapivsky et al. (2010), Taylor polynomials and radial basis functions (Ranjbar et al., 2010), Laplace-Variational iterations (Hammouch and Mekkaoui, 2010) and statistical methods based on a thermodynamic approach (Matsoukas, 2015, 2016). Asymptotic solutions have been given by van Dongen and Ernst (1988) and by Hayakawa (1987) for coagulation in the presence of source and sink terms. Extensions to bicomponent aggregation have been given by Lushnikov (1976), Matsoukas et al. (2006) and Fernández-Díaz and Gómez-García (2007, 2010). A related problem of interest is simultaneous aggregation and fragmentation that exhibits interesting dynamics, including steady state solutions (Vigil and Ziff, 1989), stationary solutions that obey detailed balance Durrett et al. (1999); Vigil (2009) or even oscillatory behavior (Matveev et al., 2017; Connaughton et al., 2018; Brilliantov et al., 2018).

Though not many new solutions have appeared in recent years, there is continuing interest in developing solution methodologies that are general and not specific to form of the aggregation kernel. The method of homotopy perturbation was recently developed to obtain analytical solutions from differential and integral equations and has been successfully used to solve a wide range of dynamical problems (Ganji, 2006; He, 2003a; Shahed, 2011). The purpose of this paper is to formulate the homotopy perturbation method (HPM) in a form appropriate for the fragmentation and aggregation equations, apply it to obtain new analytical solutions in fragmentation and aggregation, and demonstrate that the HPM provides a unified mathematical framework for population balance problems. The paper is organized as follows. In section 2 we introduce the general methodology of HPM. In section 3, HPM is adapted to the fragmentation equation and is applied to a number of standard models and present a new solution for the transient behavior of a quaternary breakup model. In section 4 we formulate the HPM method in aggregation, demonstrate the solution for several known cases (constant, sum and product kernel) and present a new solution for a nonlinear additive kernel (Ruckenstein and Pulvermacher, 1973).

## 2 The Homotopy Perturbation Method

The homotopy perturbation method (HPM) has been studied by many as a method for solving linear and nonlinear problems (He, 1999, 2000, 2003b, 2006; Nazari-Golshan et al., 2013; El-Shahed, 2005; Ganji, 2006). The HPM yields a very rapid convergence of the solution series in most cases, usually only few iterations leading to very accurate solutions. To illustrate the HPM, we consider the following differential equation

$$T(c) - h(r) = 0, \quad r \in \Omega, \quad (2.1)$$

with boundary conditions

$$B(c, \partial c / \partial n) = 0, \quad r \in \partial \Omega, \quad (2.2)$$

where  $T$  is a general differential operator and  $B$  is a boundary operator. Usually the operator  $T$  can be decomposed into two parts, a linear operator  $L$  and a nonlinear operator  $N$ , and expressed as

$$L(c) + N(c) - h(r) = 0. \quad (2.3)$$

NOTE: If the equation is linear then the non-linear part will be zero, i.e.,  $N(c) = 0$ .

HPM constructs a homotopy that satisfies

$$H[v(r, p)] = (1 - p)[L(v(r, p)) - L(c_0)] + p[T[v(r, p)] - h(r)] = 0, \quad (2.4)$$

where  $c_0$  is an initial guess to exact solution of (2.1). When  $p = 0$  then  $L(v(r, 0)) = L(c_0) = 0$ , and when  $p = 1$ , then  $T(v(r, 1)) - h(r) = 0$ . As the embedding parameter  $p$  increases monotonically from zero to unity,  $v(r, p)$  correspondingly changes from  $c_0(r)$  to  $c(r)$ . This is called deformation, and functions  $L(v) - L(c_0)$  and  $T(v) - h(r)$  are called homotopic in topology.

According to the HPM, we can first view the embedding parameter  $p$  as a small parameter, and construct the solution as a power series in  $p$ , as

$$c = \sum_{k=0}^{\infty} p^k v_k = v_0 + p v_1 + p^2 v_2 + \dots \quad (2.5)$$

Substituting (2.5) in (2.4) and then letting  $p = 1$ , we obtain the solution as

$$f = \lim_{p \rightarrow 1} c = \sum_{k=0}^{\infty} v_k. \quad (2.6)$$

The series (2.6) is a convergent for most of the cases and the rate of convergence depends on the nature of the problem He (2000). The condition for the convergence of HPM Ayati and Biazar (2015) is given as

**Theorem 2.1.** *Let  $B$  be Banach space. Then,  $\sum_{i=0}^{\infty} v_i$  converges to  $f \in B$ , if  $\exists (0 \leq \lambda < 1)$  such that  $\forall n \in N \Rightarrow \|v_n\| \leq \lambda \|v_{n-1}\|$ .*

In the following HPM will be implemented to the aggregation and fragmentation equations. It can be noticed clearly that the breakage equation is linear whereas aggregation equation is non-linear. Therefore, for solving breakage equation, HPM for linear equation will be implemented and for solving aggregation equation, HPM for non-linear equation will be implemented.

## 3 Implementation of HPM to Fragmentation

To apply HPM to fragmentation we first express the population balance equation in the integral form,

$$\frac{\partial c(m, t)}{\partial t} = -a(m)c(m, t) + \int_m^{\infty} k(m|n)a(n)c(n, t)dn, \quad (3.1)$$

where  $c_0(m) = c(m, 0)$  is the size distribution at time zero. Next we introduce a new function  $c = c(m, t; p)$  of time, size,  $p$  and define the homotopy of Eq. (3.1) as follows:

$$(1-p) \left( \frac{\partial c(m, t)}{\partial t} - \frac{\partial c(m, 0)}{\partial t} \right) + p \left\{ \frac{\partial c(m, t)}{\partial t} + a(m)c(m, t) - \int_m^{\infty} k(m|n)a(n)c(n, t)dn \right\} = 0. \quad (3.2)$$

With  $p = 0$  we obtain the initial condition,  $c(m, 0) = c_0$ ; with  $p = 1$  we obtain the complete solution. Thus by continuously varying  $p$  from 0 to 1 we obtain a continuous transformation of  $c(m, t; p)$  from the initial distribution  $c_0$  to the actual distribution  $c(m, t)$  at time  $t$ . Following HPM, we express  $c(m, t; p)$  as a power series in  $p$ ,

$$c(m, t; p) = \sum_{k=0}^{\infty} c_k(m, t)p^k, \quad (3.3)$$

where the coefficients  $c_k$  are function of time and size to be determined. According to the homotopy in Eq. (3.2) the solution to the fragmentation equation is obtained by setting  $p = 1$  in the above series:

$$c(m, t) = \lim_{p \rightarrow 1} c(m, t; p) = \sum_{k=0}^{\infty} c_k(m, t). \quad (3.4)$$

The series is convergent and the rate of convergence depends on the nature of Eq. (3.1). To obtain the coefficients  $c_k$  we insert the series (3.3) into (3.2):

$$(1-p) \left( \frac{\partial}{\partial t} \sum_{k=0}^{\infty} p^k c_k - \frac{\partial c_0}{\partial t} \right) + p \left\{ \frac{\partial}{\partial t} \sum_{k=0}^{\infty} p^k c_k + a(m) \left( \sum_{k=0}^{\infty} p^k c_k \right) - \int_m^{\infty} k(m|n)a(n) \left( \sum_{k=0}^{\infty} p^k c_k \right) dn \right\} = 0, \quad (3.5)$$

collect terms in powers of  $p$  and set their coefficients to zero. We obtain:

$$p^0 : \quad c_0 = c(m, 0), \quad (3.6)$$

$$p^1 : \quad \frac{\partial c_1}{\partial t} = -a(m)c_0(m, t) + \int_m^{\infty} k(m|n)a(n)c_0(n, t)dn, \quad (3.7)$$

⋮

$$p^k : \quad \frac{\partial c_k}{\partial t} = -a(m)c_{k-1}(m, t) + \int_m^{\infty} k(m|n)a(n)c_{k-1}(n, t)dn. \quad (3.8)$$

The first term,  $c_0$ , is equal to the initial distribution and each subsequent term involves the coefficient of previous order. This produces a closed recursion for  $c_k$ . The  $n$ th order approximation of the solution, is obtained by truncating the series past the  $n$ th term. In several cases we will be able to obtain closed form solutions for the infinite sum. Below we first reproduce known solutions to the fragmentation equation using HPM, and then obtain new results for a case that has not been previously reported in literature.

### 3.1 Case I: Random fragmentation with linear selection function

In random fragmentation with linear selection function two fragments are formed and their distribution is given by

$$k(m|n) = \frac{2}{n}, \quad (3.9)$$

and the fragmentation rate is

$$a(m) = m. \quad (3.10)$$

The general form of the factors  $c_k(m, t)$  is given by Eq. (3.8), which now becomes

$$\frac{\partial c_k(m, t)}{\partial t} = x c_{k-1}(m, t) - 2 \int_0^\infty \frac{c_{k-1}(n, t)}{n} dn. \quad (3.11)$$

Next we obtain the solution for monodisperse and exponential initial distribution.

### 3.1.1 Monodisperse initial conditions

We start with  $c_0 = f_0 = \delta(m - 1)$  and obtain the functions  $c_k$  recursively:

$$c_0 = \delta(m) \quad (3.12)$$

$$c_1(m, t) = 2t\theta(1 - m) - tx\delta(m - 1) \quad (3.13)$$

$\vdots$

$$c_k(m, t) = \frac{t^2 (1 - m) (-tm)^{k-2}}{(k - 2)!} \theta(1 - m), \quad (3.14)$$

where  $\theta$  is the Heaviside step function. The distribution is given by the infinite series

$$c(m, t) = \sum_{k=0}^{\infty} \frac{t^2 (1 - m) (-tm)^{k-2}}{(k - 2)!} \theta(1 - m). \quad (3.15)$$

As shown in the Appendix, this is equal to

$$c(m, t) = e^{-tm} (\delta(m - 1) + \theta(a - m) (2t + t^2(1 - m))). \quad (3.16)$$

This is the same as the result given by Ziff and McGrady (Ziff and McGrady, 1985).

### 3.1.2 Exponential initial condition

Starting with  $c_0(m, t) = f_0(m, t) = e^{-m}$ , the  $c_k$ 's are

$$c_0(m, t) = e^{-m}, \quad (3.17)$$

$$c_1(m, t) = t (-e^{-m}) (m - 2), \quad (3.18)$$

$\vdots$

$$c_k(m, t) = \frac{(-m)^k t^{k-2} (-2kt + (k - 1)k + t^2)}{k!} e^{-t}. \quad (3.19)$$

The size distribution in this case is given by

$$c(m, t) = \sum_{k=0}^{\infty} \frac{(-m)^k t^{k-2} (-2kt + (k - 1)k + t^2)}{k!} e^{-t} \rightarrow (1 + t)^2 e^{-m(1+t)}. \quad (3.20)$$

This is the same as the solution given by Ziff and McGrady (Ziff and McGrady, 1985).

## 3.2 Case II: Random fragmentation with quadratic selection function

In this model the breakup rate is a quadratic function of size,  $a(m) = m^2$  and the fragment size distribution is again given by  $\beta(m|n) = 2/n$ . The initial condition is exponential. The general recursion for  $c_k(m, t)$  is

$$\frac{\partial c_k(m, t)}{\partial t} = \int_0^t m^2 c_{k-1}(m, t) - 2 \int_0^\infty \frac{c_{k-1}(n, t)}{n^2} dn. \quad (3.21)$$

We obtain solutions for monodisperse and exponential initial conditions:

### 3.2.1 Monodisperse initial condition

With  $c_0(m, t) = \delta(m - 1)$  the functions  $c_k$  are:

$$c_0(m, t) = \delta(m - 1), \quad (3.22)$$

$$c_1(m, t) = 2t\theta(1 - m) - tx^2\delta(m - 1), \quad (3.23)$$

$\vdots$

$$c_k(m, t) = \frac{\delta(t - 1)(-t^2x)^k}{k!} + \frac{2m\theta(1 - t)(-t^2m)^{k-1}}{(k - 1)!}. \quad (3.24)$$

The size distribution is

$$c(m, t) = \sum_{k=0}^{\infty} \left\{ \frac{\delta(t - 1)(-t^2x)^k}{k!} + \frac{2x\theta(1 - t)(-t^2x)^{k-1}}{(k - 1)!} \right\} \rightarrow e^{-tm^2} (\delta(m - 1) + 2at\theta(1 - m)). \quad (3.25)$$

This again agrees with the solution of Ziff and McGrady ([Ziff and McGrady, 1985](#)).

### 3.2.2 Exponential initial condition

Starting with  $c_0(m) = e^{-m}$  and solving recursively we obtain

$$c_k(m, t) = \frac{(-t)^k m^{2k-2}}{k!} (-2km - 2k + m^2) e^{-m}. \quad (3.26)$$

The solution is constructed by computing the infinite series,

$$c(m, t) = \sum_{k=0}^{\infty} \frac{(-t)^k m^{2k-2}}{k!} (-2km - 2k + m^2) e^{-m} \rightarrow e^{-tm^2} (\delta(m - 1) + 2t\theta(1 - m)). \quad (3.27)$$

This is the same as the result obtained by Ziff and McGrady ([Ziff and McGrady, 1985](#)).

## 3.3 Case III: Fragmentation with power-law rate

In this model the selection function is  $a(m) = m^\alpha$  and the fragment size distribution is given by

$$k(m|n) = \frac{\alpha}{n} \left(\frac{m}{n}\right)^{\alpha-2}, \quad (3.28)$$

which corresponds to mean number of fragments

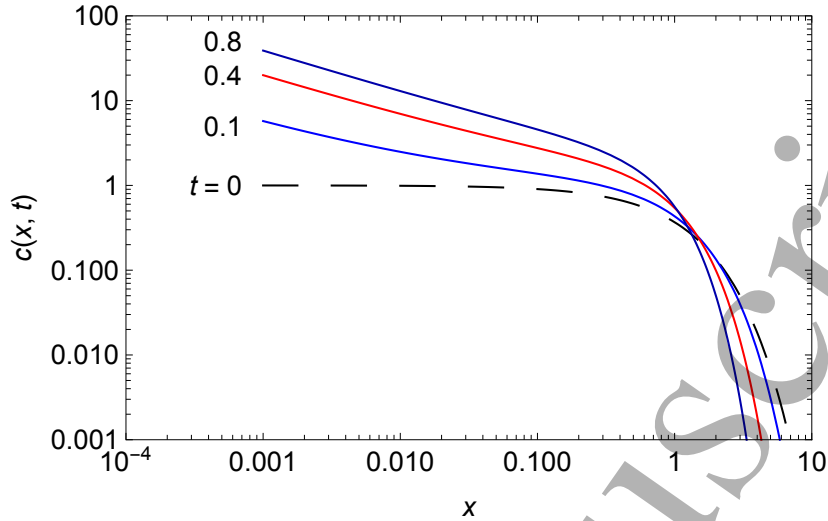
$$\bar{f} = \frac{\alpha}{\alpha - 1}. \quad (3.29)$$

By varying  $\alpha$  between 1 and 2 the number of fragments ranges from  $\infty$  to 2. With  $\alpha = 2$ , in particular, this model defaults to the random distribution of fragments with quadratic breakage rate. Equation (3.8) in this model now is

$$\frac{\partial c_k(m, t)}{\partial t} = x^\alpha c_{k-1}(m, t) - \alpha \int_m^\infty nm^{\alpha-2} c_{k-1}(n, t) dn. \quad (3.30)$$

Using  $c_0(m, t) = e^{-m}$  the functions  $c_k$  are

$$c_k(m, t) = \frac{(-t)^k m^{\alpha k-2}}{k!} (-\alpha k - \alpha km + m^2) e^{-m}. \quad (3.31)$$



**Figure 1** Evolution of the size distribution for fragmentation with power-law rate. The distribution is calculated from Eq. (3.32) with  $\alpha = 3/2$ .

Inserting this result into the infinite series in Eq. (3.4) we obtain

$$c(m, t) = \sum_{k=0}^{\infty} \frac{(-t)^k m^{\alpha k - 2}}{k!} (-\alpha k - \alpha k m + m^2) e^{-m} \rightarrow e^{-m - tm^\alpha} (1 + \alpha t(m^{\alpha-2} + m^{\alpha-1})), \quad (3.32)$$

which gives the size distribution at all times. This distribution is plotted in Fig. 1 for the case  $\alpha = 3/2$ .

This model has been studied in the literature (Montroll and Simha, 1940; Austin and Luckie, 1972; Klimpel and Austin, 1977) and it is known to reach a scaling solution (Ziff and McGrady, 1985), its time-dependent solution however has not been previously obtained in the literature and is a new result. The HPM solution obeys the scaling derived by Ziff and McGrady (Ziff and McGrady, 1985), as can be easily demonstrated. Dividing Eq. (3.16) by  $m^2/\alpha$  and setting  $m = (z/t)^{1/\alpha}$  we find

$$\frac{\alpha c(m, t)}{m^2} = \frac{1}{\alpha} \left\{ t^{-2/\alpha} e^{-t^{-1/\alpha} (-z^{1/\alpha}) - z} \left( \alpha t^{1/\alpha} z^{(\alpha+1)/\alpha} + \alpha z t^{2/\alpha} + z^{2/\alpha} \right) \right\}, \quad (3.33)$$

whose limit at long  $t$  is

$$\frac{\alpha c(m, t)}{m^2} \rightarrow z e^{-z} \equiv \Phi(z), \quad (3.34)$$

and  $\Phi(z)$  is the scaling function. The scaling limit of the distribution is

$$c(m, t) \sim \alpha m^2 \Phi(tm^\alpha). \quad (3.35)$$

The scaling function given by Ziff and McGrady is  $\Phi(z) = z^{\gamma/\alpha} e^{-z} \Gamma(\gamma/\alpha)$  (the Filippov model in Table 1 of Ref. (Ziff and McGrady, 1985)), which for  $\gamma = \alpha$  reduces to the result obtained here.

## 4 Application of the HPM to Aggregation

We construct the homotopy of equation (1.4) as follows

$$(1-p) \left( \frac{\partial c(m,t)}{\partial t} - \frac{\partial c_0(m,t)}{\partial t} \right) + p \left( \frac{\partial c(m,t)}{\partial t} - \frac{1}{2} \int_0^m \beta(m-n,n)c(m-n,t)c(n,t)dn + \int_0^\infty \beta(m,n)c(m,t)c(n,t)dn \right) = 0 \quad (4.1)$$

$$\frac{\partial c(m,t)}{\partial t} - (1-p) \frac{\partial c_0(m,t)}{\partial t} + p \left( -\frac{1}{2} \int_0^m \beta(m-n,n)c(m-n,t)c(n,t)dn + \int_0^\infty \beta(m,n)c(m,t)c(n,t)dn \right) = 0. \quad (4.2)$$

We set

$$c = \sum_{k=0}^{\infty} p^k c_k \quad (4.3)$$

in the above equation and equate the coefficients of  $p^k$  on both sides of the equation. The result is a recursion for the coefficients  $c_k$ :

$$c_0(m,t) = c(m,0), \quad (4.4)$$

$$\frac{\partial c_k(m,t)}{\partial t} - \frac{1}{2} \int_0^m \beta(m-n,n) \left( \sum_{l=0}^{k-1} c_l(m-n,t)c_{k-l-1}(n,t) \right) dn + \int_0^\infty \beta(m,n) \left( \sum_{l=0}^{k-1} c_l(n,t)c_{k-l-1}(m,t) \right) dn = 0; \quad k = 1, 2, \dots \quad (4.5)$$

Next we obtain analytical solution for various aggregation kernels and initial conditions.

### 4.1 Constant aggregation kernel $\beta(m,n) = 1$ , exponential initial condition

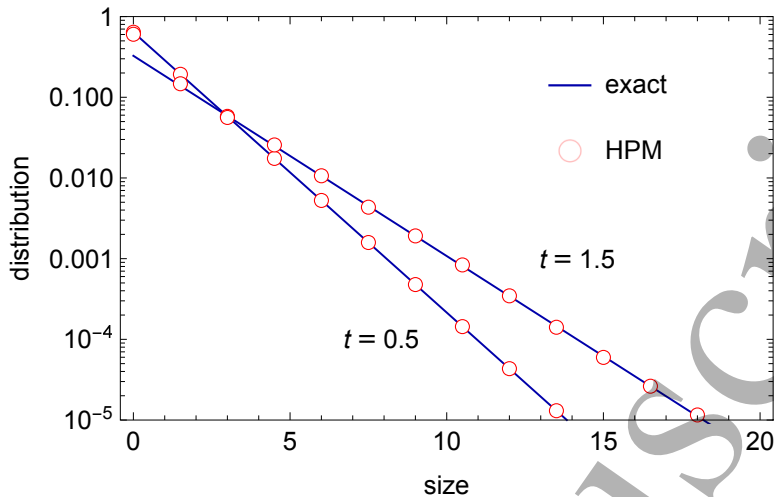
The general form of the factors  $c_k(m,t)$  is given by Eq. (4.5), which

$$\frac{\partial c_k(m,t)}{\partial t} - \frac{1}{2} \int_0^m \left( \sum_{l=0}^{k-1} c_l(m-n,t)c_{k-l-1}(n,t) \right) dn + \int_0^\infty \left( \sum_{l=0}^{k-1} c_{k-l-1}(m,t)c_l(n,t) \right) dn = 0; \quad k = 1, 2, 3, \dots \quad (4.6)$$

The solution will be obtained for exponential initial condition, i.e.,  $c_0(m) = e^{-m}$ . So, let us initialize the solution with  $c_0 = e^{-m}$  and obtain the functions  $c_k$  recursively as follows.

$$c_0 = e^{-m}, \quad (4.7)$$

$$c_1 = \frac{t}{2^1 1!} (m-2) e^{-m}, \quad (4.8)$$



**Figure 2** Size distribution for constant kernel with exponential initial condition at  $t = 0.5$  and  $t = 1.5$ . The HPM solution in this case is exact.

$$c_2 = \frac{t^2}{2^2 2!} (m^2 - 6m + 6) e^{-m}, \quad (4.9)$$

⋮

$$c_k = e^{-m} \frac{(k+1)! t^k}{2^k} \sum_{r=0}^k \frac{(-1)^r m^{k-r}}{r!(k-r+1)!(k-r)!}. \quad (4.10)$$

The partial sum of the series solution is obtained as

$$c(m, t) \approx \phi_n(t, m) = \sum_{k=0}^n c_k. \quad (4.11)$$

In the limit  $n \rightarrow \infty$  we obtain

$$c(m, t) = e^{-m} \sum_{k=0}^{\infty} \frac{t^k}{2^k k!} \text{HypergeometricU}[-k, 2, m]. \quad (4.12)$$

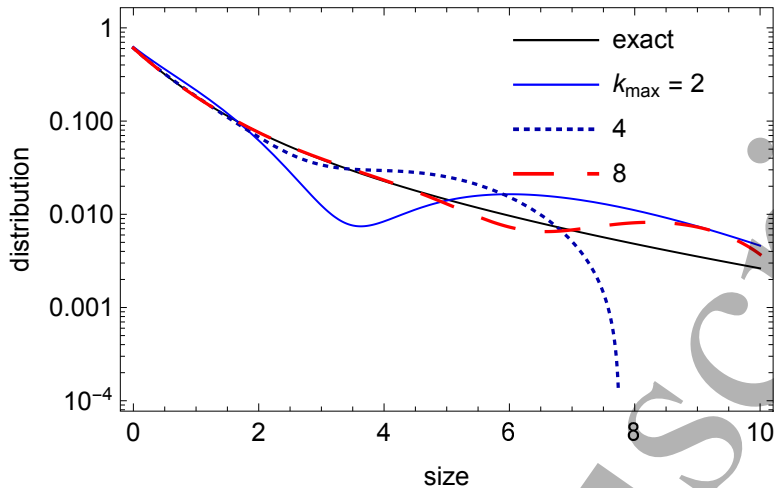
Here,  $\text{HypergeometricU}[x, y, z]$  is confluent hypergeometric function  $U[x, y, z]$ , as implemented in Mathematica, and defined as

$$\text{HypergeometricU}[x, y, z] = \frac{1}{\Gamma(x)} \int_0^{\infty} e^{-zt} t^{x-1} (1+t)^{y-x-1} dt. \quad (4.13)$$

The known solution in this case is

$$c(m, t) = \left( \frac{2}{t+2} \right)^2 e^{-2m/(t+2)}, \quad (4.14)$$

as in [Ranjbar et al. \(2010\)](#). Eqs. (4.11) and (4.14) are equivalent for  $t < 2$ . We demonstrate this by a graphical comparison in Fig. 2), which shows the distribution at two different times and demonstrates that it remains exponential.



**Figure 3** HPM solution of the sum kernel with exponential initial condition at  $t = 0.5$  using 2, 4 and 8 terms. The exact solution is Eq. (4.15).

## 4.2 Sum aggregation kernel $\beta(m, n) = m + n$

For the sum kernel with exponential initial condition,  $c_0(m) = e^{-m}$ , the analytical solution is Kumar (2006)

$$c(m, t) = \frac{(1 - \tau)e^{-(1+\tau)m}}{m\sqrt{\tau}} I_1[2m\sqrt{\tau}], \quad (4.15)$$

where  $\tau = 1 - e^{-t}$  and  $I_1[m]$  is the modified Bessel function of the first kind. The general form of the factors  $c_k(m, t)$  is given by Eq. (4.5), which

$$\begin{aligned} \frac{\partial c_k}{\partial t} = & \frac{1}{2} \int_0^m m \left( \sum_{l=0}^{k-1} c_l(m-n, t) c_{k-1-l}(n, t) \right) dn \\ & - \int_0^\infty (m+n) \left( \sum_{l=0}^{k-1} c_{k-1-l}(m, t) c_l(n, t) \right) dn, \quad k = 1, 2, 3... \end{aligned} \quad (4.16)$$

We initialize with  $c_0 = e^{-m}$  and obtain the functions  $c_k$  recursively. The terms up to order 4 are shown below:

$$c_0 = e^{-m}, \quad (4.17)$$

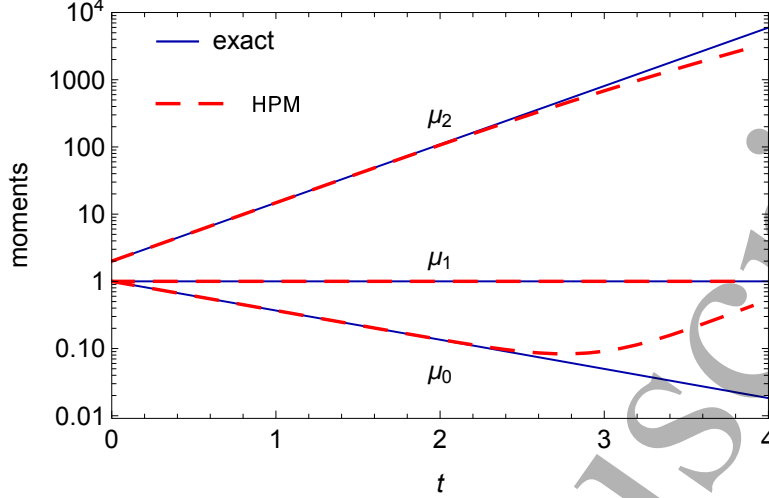
$$c_1 = \frac{e^{-m}t}{2} (-2 - 2m + m^2), \quad (4.18)$$

$$c_2 = \frac{e^{-m}t^2}{12} (6 + 18m - 3m^2 - 6m^3 + m^4), \quad (4.19)$$

$$c_3 = \frac{e^{-m}t^3}{144} (-24 - 168m - 60m^2 + 120m^3 + 12m^4 - 12m^5 + m^6), \quad (4.20)$$

$$c_4 = \frac{e^{-m}t^3}{2880} (120 + 1800m + 2100m^2 - 1800m^3 - 1180m^4 + 360m^5 + 70m^6 - 20m^7 + m^8). \quad (4.21)$$

Figure 3 shows the comparison between HPM, constructed using 2, 4 and 8 terms, and the literature result in Eq. (4.15). As this figure shows, increasing the order of the approximation improves the accuracy of the HPM



**Figure 4** Moments of the sum kernel calculated using  $k_{\max} = 8$  in the HPM.

solution to larger sizes. Figure 4 shows the three lower moments calculated with the 8-term approximation. The moments of the theoretical solution are

$$\mu_0 = e^{-t}, \quad \mu_1 = 1, \quad \mu_2 = 2e^{2t} \quad (4.22)$$

in this case the truncated HPM is the corresponding power series up to the truncated term. With eight terms the truncation is accurate up to  $t \approx 2$ .

### 4.3 Product aggregation kernel $\beta(m, n) = mn$

Under this aggressive kernel newly formed particles aggregate at a higher rate than their parents and the system exhibits gelation (Ernst et al., 1984). The solutions obtained here are valid only before the gel point. The general form of the factors  $c_k(m, t)$  is given by Eq. (4.5), which now becomes

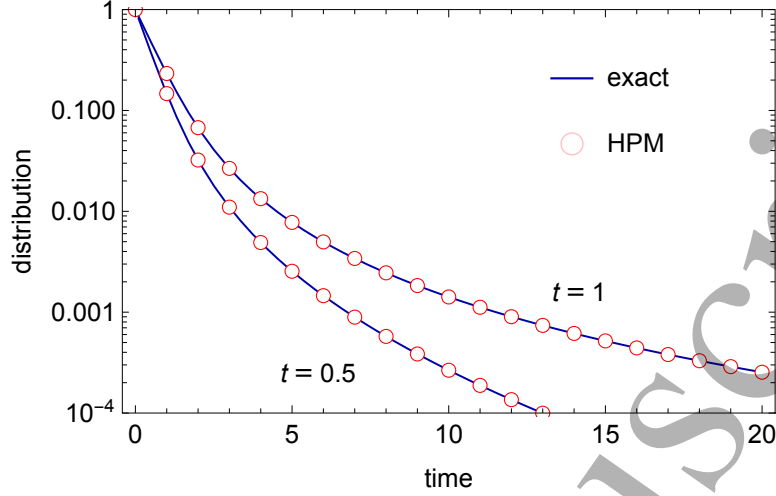
$$\begin{aligned} \frac{\partial c_k(m, t)}{\partial t} - \frac{1}{2} \int_0^m (m-n)n \left( \sum_{l=0}^{k-1} c_l(m-n, t) c_{k-1-l}(n, t) \right) dn \\ + \int_0^\infty mn \left( \sum_{l=0}^{k-1} c_{k-1-l}(m, t) c_l(n, t) \right) dn; \quad k = 1, 2, 3... \end{aligned} \quad (4.23)$$

The solution will be obtained for two different initial conditions, i.e., for  $c_0(m) = e^{-m}$  and  $c_0(m) = \frac{e^{-m}}{m}$ .

#### 4.3.1 Exponential initial condition, $c_0(m) = e^{-m}$

Its analytic solution follows Ernst et al. (1984) as

$$c(m, t) = \sum_{k=0}^{\infty} e^{-(t+1)m} \frac{t^k m^{3k}}{(k+1)! \Gamma(2k+2)}. \quad (4.24)$$



**Figure 5** HPM solution for the product kernel with exponential initial conditions at  $t = 0.5$  and  $t = 1$ . The exact solution is Eq. (4.24).

We start with  $c_0 = e^{-m}$  and obtain the functions  $c_k$  recursively

$$c_0 = e^{-m}, \quad (4.25)$$

$$c_1 = \frac{(tm)^1}{2!3!} (-12 + m^2) e^{-m}, \quad (4.26)$$

$$c_2 = \frac{(tm)^2}{3!5!} (360 - 60m^2 + m^4) e^{-m}, \quad (4.27)$$

$\vdots$

$$c_k = 2e^{-m} (mt)^k \sum_{r=0}^k \frac{(-1)^r m^{2k-2r}}{r!(k-r)!(2k-2r+2)!}. \quad (4.28)$$

The partial sum of the series solution is defined obtained as

$$c(m, t) \approx \phi_n(t, m) = \sum_{k=0}^n c_k. \quad (4.29)$$

The solution calculated by HPM is compared with the exact solution (4.24) in Fig. 5 using 100 for both the HPM summation and the literature result in Eq. (4.24). The agreement is excellent.

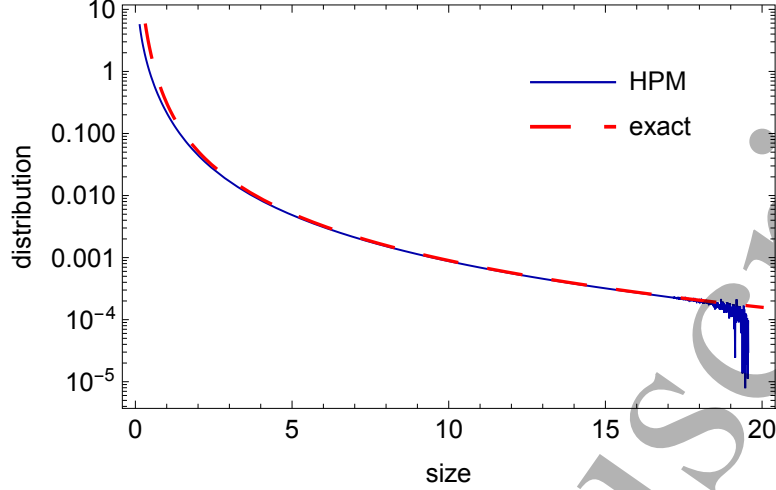
#### 4.3.2 Initial condition $c_0(m) = e^{-m}/m$

The analytical solution can be found in [Ranjbar et al. \(2010\)](#)

$$c(m, t) = \frac{I[2m\sqrt{t}]}{m^2\sqrt{t}} e^{-Tm}, \quad T = \begin{cases} 1+t, & t \leq 1, \\ 2\sqrt{t}, & \text{otherwise.} \end{cases} \quad (4.30)$$

Here  $I$  is the modified Bessel's first kind function:

$$I[m] = \frac{1}{\pi} \int_0^{\pi} e^{(m \cos \theta)} \cos \theta d\theta. \quad (4.31)$$



**Figure 6** HPM solution for the product kernel with initial condition  $c_0(m) = e^{-m}/m$  at  $t = 1$ . compared with exact solution from Eq. (4.30).

Initialize the approximation with  $c_0 = \frac{e^{-m}}{m}$  and obtain the functions  $c_k$  recursively as follows:

$$c_0 = \frac{e^{-m} m^{-1}}{1}, \quad (4.32)$$

$$c_1 = \frac{t}{2(1!)^2} (m-2) e^{-m}, \quad (4.33)$$

$$c_2 = \frac{t^2 m}{3(2!)^2} (m^2 - 6m + 6) e^{-m}, \quad (4.34)$$

$$\vdots \quad (4.35)$$

$$c_k = e^{-m} m^{k-1} t^k \sum_{r=0}^k \frac{(-1)^r m^{k-r}}{r!(k-r+1)!(k-r)!}. \quad (4.36)$$

Similar to constant kernel case, the partial sum of the series solution for the product kernel can be obtained as

$$c(m, t) \approx \phi_n(m, t) = \sum_{k=0}^n c_k. \quad (4.37)$$

As  $n \rightarrow \infty$ , we obtain

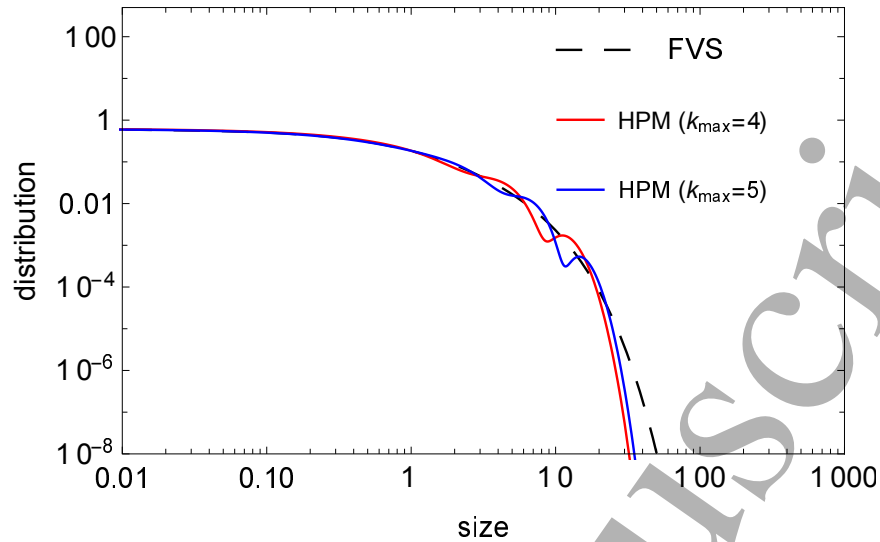
$$e^{-m} \sum_{k=0}^{\infty} \frac{t^k m^{k-1}}{k!(k+1)!} \text{Hypergeometric}U[-k, 2, m]. \quad (4.38)$$

The agreement is very good (see Fig. 6) except at very large  $m$  due to numerical precision. We note, however that the value of the distribution at this point is approximately 5 decades below its maximum and that this loss of accuracy has no discernible effect in the calculation of the lower moments.

#### 4.4 Ruckenstein/Pulvermacher kernel $\beta(m, n) = m^{2/3} + n^{2/3}$

We now obtain the analytical solution of the size distribution for the kernel

$$\beta(m, n) = m^{2/3} + n^{2/3}. \quad (4.39)$$



**Figure 7** The distribution for the Ruckenstein/Pulvermacher kernel with exponential initial conditions. Results for  $t = 0.4$  using 5 terms in the truncated series.

This kernel was used to describe the process of particle migration and coalescence on a heated substrate and has been proposed as a model for the aging of supported metal catalysts (Ruckenstein and Pulvermacher, 1973). Here we obtain the solution for exponential initial condition  $c_0(m) = e^{-m}$ . The general form of the factors  $c_k(x, t)$  is given by Eq. (4.5) and the first three terms are given below:

$$c_0 = e^{-m}, \quad (4.40)$$

$$c_1 = \frac{e^{-m}t}{5}[-5m^{2/3} + 3m^{5/3} - 5\Gamma(5/3)], \quad (4.41)$$

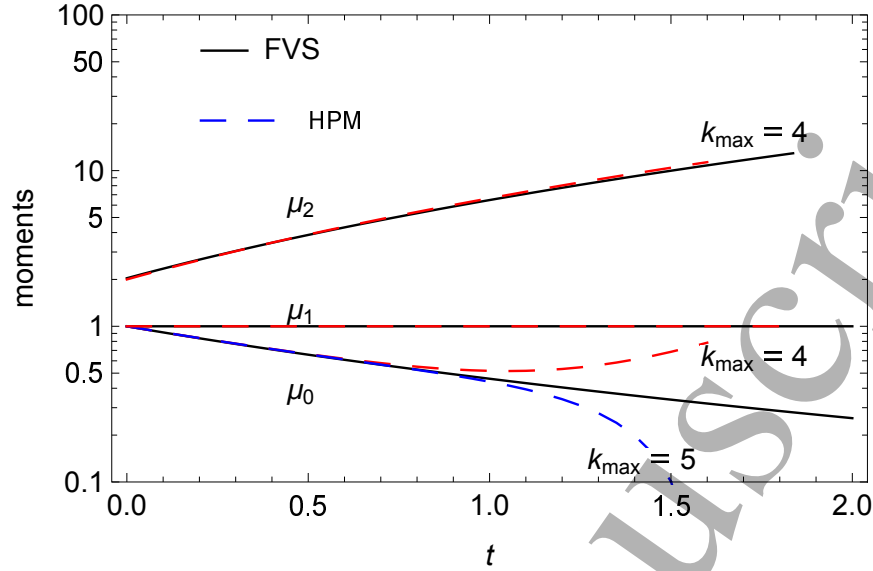
$$c_2 = \frac{e^{-m}t^2}{50400\Gamma(13/6)}[315 \times 2^{2/3}\sqrt{\pi}m^{7/3}(-10 + 3m)\Gamma(5/3) + 8(3150m^{4/3} - 3240m^{7/3} + 567m^{10/3} + 6300m^{2/3}\Gamma(2/3) - 560(\Gamma(1/3) - 5\Gamma(2/3)^2) - 1260m^{5/3}(\Gamma(2/3) + 3\Gamma(5/3)))\Gamma(13/6)]. \quad (4.42)$$

Higher order terms become increasingly more complex but they may be calculated systematically by recursive application of Eq. (4.5).

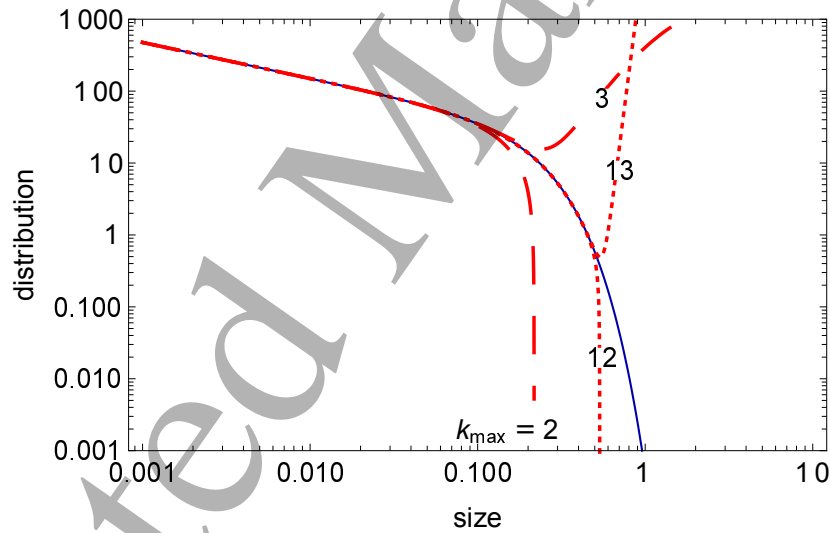
## 5 Discussion

The HPM expresses the solution of the coagulation and fragmentation equations as a series expansion in terms of a set of functions  $c_k(m, t)$  that are calculate recursively starting with the initial solution  $c_0(0, t) = c(m, t = 0)$ . Recursive series expansions are not new in population balances. Melzak (1957a,b) expressed the solution as a power series in time with coefficients that depend only on the size of the clusters, and obtained these coefficients by a similar recursion. Lushnikov (Lushnikov, 1973) obtained these coefficients in closed form in the special case of the constant and sum kernels under monodisperse initial conditions. The same expansion was used by Song and Poland (1992) to obtain asymptotic solutions for the moments of the coagulation equation. The HPM formulation has the advantage that it is not limited to monodisperse initial conditions and provides a systematic methodology that summarizes all known results. It is interesting to examine the degree to which finite truncations may produce accurate approximations of the full solution.

Figure 3 illustrates the effect of the order of the truncation for the product kernel with exponential initial distribution. The truncated approximations represent the small clusters quite well but become oscillatory at larger sizes. Increasing the order of the approximation extends the range of accuracy but at sufficiently



**Figure 8** Lower moments for the Ruckenstein/Pulvermacher kernel. HPM solutions are constructed with truncations to  $k_{\max} = 4$  and 5.



**Figure 9** Truncated series to  $k_{\max}$  for  $\alpha = 3/2$ ,  $t = 10$ . The dashed lines are truncated approximations of the full solution (solid line).

large sizes the approximation regains the oscillatory behavior. On the other hand, the lower moments of the distribution exhibit remarkable stability. As Figure 4 shows, the series truncated to  $k_{\max} = 8$  gives excellent agreement for the zeroth and first order moments, even at later times when the oscillatory behavior in the distribution is even stronger than that seen in Fig. 3. In the case of the Ruckenstein/Pulvermacher kernel the oscillatory behavior in the distribution is still present (Fig. 7) but less severe. The zeroth moment is predicted accurately at short times and the accuracy improves with increasing number of terms (Fig. 8). The first moment on the other hand is conserved even when the zeroth moment breaks down. The second moment on the other hand is predicted with higher accuracy even after the truncated HPM begins to fail for the zeroth order moment.

The situation in breakup is different. Figure 9 compares truncated approximations of the full solutions for

the breakup kernel in Eq. (3.28) with  $\alpha = 3/2$  at  $t = 10$  and  $k_{\max} = 2, 3, 12$  and  $13$  ( $k_{\max}$  is the maximum order of the term retained in the series). As with aggregation, the accuracy is very good at small size but breaks at larger sizes. However, deviations are not oscillatory and truncations with even  $k_{\max}$  produce stable distributions whose large size tail decays to zero. Truncations are in excellent agreement up to a maximum mass, whose value increases with the order of the truncation. In this particular case, even a very low order approximation such as  $k_{\max} = 2$  represents the distribution very well over two decades in  $c(m)$ .

## 6 Conclusions

The HPM provides a systematic methodology to solve for the size distribution in aggregation and fragmentation processes with arbitrary initial conditions. The method was shown to reproduce previously known solutions and was used to produce two new results, for the power-law kernel in fragmentation and the Ruckenstein/Pulvermacher kernel in aggregation. The method offers the possibility of constructing approximations based on truncations of the HPM series. The accuracy of truncated approximations is limited to smaller sizes. In the case of fragmentation, which proceeds in the direction of smaller sizes, HPM approximations are remarkably accurate, even when the distribution diverges at zero size. Aggregation, on the other hand, produces ever larger sizes and this limits the practical usefulness of the truncated solutions.

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