Solutions and critical times for the polydisperse coagulation equation when a(x,y)=A+B(x+y)+Cxy

1983 J. Phys. A: Math. Gen. 16 3127

(http://iopscience.iop.org/0305-4470/16/13/037)

J. Phys. A: Math. Gen. 16 (1983) 3127–3132. Printed in Great Britain

Solutions and critical times for the polydisperse coagulation equation when $a(x, y) = A + B(x + y) + Cxy$

John L Spouge
Trinity College, Oxford OX1 3BH, UK

Received 4 January 1983

Abstract. This paper gives solutions of the polydisperse Smoluchowski coagulation equation

$$\partial f(x, t)/\partial t = \tfrac{1}{2}\int_0^x f(y, t)f(x-y, t)a(y, x-y)\,\mathrm{d}y - f(x, t)\int_0^\infty f(y, t)a(x, y)\,\mathrm{d}y,$$

$$f(x, 0) = g(x)$$

for arbitrary $g(x)$ when the coagulation kernel

$$a(x, y) = A + B(x + y) + Cxy.$$

The solutions are given as recursions and infinite series and are practical for computation. For the given kernels we also give the gelation times t_g at which

$$M_2(t) = \int_0^\infty x^2 f(x, t)\,\mathrm{d}x$$

becomes infinite.

1. Introduction

The polydisperse Smoluchowski coagulation equation is the partial differential equation

$$\frac{\partial f(x, t)}{\partial t} = \tfrac{1}{2}\int_0^x f(y, t)f(x-y, t)a(y, x-y)\,\mathrm{d}y - f(x, t)\int_0^\infty f(y, t)a(x, y)\,\mathrm{d}y,$$

$$f(x, 0) = g(x). \tag{1}$$

This paper gives practical solutions of (1) for arbitrary $g(x)$ when

$$a(x, y) = A + B(x + y) + Cxy. \tag{2}$$

Drake's (1972) review of coagulation solves some of these cases by Laplace transforms but the solutions are not always practical for computation.

Equation (1) and related equations have been used to model clumping processes in astrophysics (Barrow 1981), meteorology (Drake 1972), polymer chemistry (Cohen and Benedek 1982), haematology (Popel *et al* 1975), colloid chemistry (Lushnikov 1973) and aerosol science (Ramabhadran *et al* 1976).

Equation (1) has the following physical interpretation: consider a fixed volume of space containing a large number of randomly moving particles. The particles vary in mass and clump irreversibly (or coagulate) when they meet. $f(x, t)$ gives the mass spectrum at time t, i.e. $f(x, t)\,dx$ is the average number of particles per unit volume having mass x to $x+dx$. (All averages will be taken over the fixed volume.) $g(x)$ is the initial mass spectrum. The volume-average number of coalescences between particles of mass x to $x+dx$ and those of mass y to $y+dy$ during the time interval t to $t+dt$ is $f(x, t)f(y, t)a(x, y)\,dx\,dy\,dt$. $a(x, y)$ is called the coagulation kernel. Equation (1), a conservation of mass relation, gives $\partial f(x, t)/\partial t$ as the difference of two terms (a) and (b).

(a) The rate of formation of x-masses by coagulation of y- and $(x-y)$-masses.

(b) The rate of disappearance of x-masses by coagulation with y-masses.

In future, we shall suppress dependence on time (e.g. $f(x)=f(x, t)$) when the meaning is clear.

The moments of the mass spectrum are

$$M_n(t)=\int_0^\infty x^n f(x)\,dx \qquad n=0, 1, 2 \ldots . \tag{3}$$

If all moments are initially finite, the divergence of any moment $M_n(t)$ heralds the appearance of an infinite mass (Ziff 1980, Leyvraz and Tschudi 1981). We call this phenomenon gelation (borrowing the term from polymer chemistry), and the time that gelation first occurs the gelation time t_g. We shall assume hereafter that all moments are initially finite.

Equation (1) usually occurs in its monodisperse form

$$\frac{dx_n}{dt}=\tfrac{1}{2}\sum_{i=1}^{n-1} x_i x_{n-i} a_{i,(n-i)} - x_n \sum_{i=1}^{\infty} x_i a_{n,i}$$

$$x_1(0)=1 \qquad x_2(0)=x_3(0)=\ldots=0. \tag{4}$$

The solutions of (1) and (4) are related by

$$f(x, t)=\sum_{n=1}^{\infty} x_n(t)\delta(x-n) \tag{5}$$

($\delta(x)$ is the Dirac delta function) and (4) is the specialisation of (1) wherein all particles are initially of unit mass.

If $a_{ij}=(ij)^w$ in (4), Ernst *et al* (1982), Ziff *et al* (1982) and Ziff (1980) have shown $t_g<\infty$ for $w>\frac{1}{2}$. Leyvraz and Tschudi (1981) give t_g for the kernels (2) when $B^2=AC$. This paper gives t_g for all the kernels (2). Ziff and Stell (1980) give post-gelation solutions of (4), some of which are easily extended to (1), but we shall confine our interest to pre-gelation solutions of (1).

Let us choose units of mass and volume so that $M_0(0)=M_1(0)=1$. For $t<t_g$, finiteness of the moments yields

$$\frac{dM_n}{dt}=\tfrac{1}{2}\int_0^\infty\int_0^\infty [(x+y)^n-x^n-y^n]f(x)f(y)a(x, y)\,dx\,dy. \tag{6}$$

Hence $M_1(t)\equiv 1$ for $t<t_g$. Hereafter we restrict ourselves to the kernels (2). $\mu=M_0(t)$ satisfies

$$d\mu/dt=-\tfrac{1}{2}(A\mu^2+2B\mu+C)=-\tfrac{1}{2}D(\mu). \tag{7}$$

Solutions of (7), subject to $\mu(0)=1$, are given in table 1.

Table 1. Solutions of $\mathrm{d}\mu/\mathrm{d}t = -\frac{1}{2}(A\mu^2 + 2B\mu + C)$.

Cases	t	μ
$C=0$: $B=0$	$\frac{2}{A}\frac{1-\mu}{\mu}$	$\frac{2}{At+2}$
: $B\neq 0$	$\frac{1}{B}\ln\frac{A\mu+2B}{\mu(A+2B)}$	$\frac{2B}{(A+2B)\,\mathrm{e}^{Bt}-A}$
$C\neq 0$: $A=0$: $B=0$	$\frac{2}{C}(1-\mu)$	$\frac{2-Ct}{2}$
: $B\neq 0$	$\frac{1}{B}\ln\frac{2B+C}{2B\mu+C}$	$\frac{(2B+C)\mathrm{e}^{-Bt}-C}{2B}$
: $A\neq 0$: $B^2=AC$	$\frac{2A(1-\mu)}{(A\mu+B)(A+B)}$	$\frac{2A-B(A+B)t}{A[2+(A+B)t]}$
: $B^2>AC$ $\mu_\pm = \frac{-B\pm(B^2-AC)^{1/2}}{A}$	$\frac{2}{A(\mu_+-\mu_-)}\ln\left(\frac{1-\mu_+}{\mu-\mu_+}\frac{\mu-\mu_-}{1-\mu_-}\right)$	$\mu_+ + \frac{(\mu_+-\mu_-)(1-\mu_+)}{(1-\mu_-)\exp[A(\mu_+-\mu_-)t/2]-(1-\mu_+)}$
: $B^2<AC$ $\alpha=-\frac{B}{A},\quad \gamma=\frac{(AC-B^2)^{1/2}}{A}$	$\frac{2}{A\gamma}\left(\tan^{-1}\frac{1-\alpha}{\gamma}-\tan^{-1}\frac{\mu-\alpha}{\gamma}\right)$	$\alpha+\gamma\frac{1-\alpha-\gamma\tan(\frac{1}{2}A\gamma t)}{\gamma+(1-\alpha)\tan(\frac{1}{2}A\gamma t)}$

To derive gelation times of (4) we note that for $n=2$, (6) becomes

$$dM_2/dt = A + 2BM_2 + CM_2^2. \tag{8}$$

Substituting $\mu_2 = 1/M_2$ and $\tau = 2t$ into (8) yields an equation of the form (7). When $t=0$, $\mu_2 = 1/M_2(0)$ and when $t = t_g$, $\mu_2 = 0$. These conditions allow us to use the solutions of (7) to solve (8) for t_g. The results are given in table 2 and are in Drake (1972, section 5.4).

Table 2. Gelation times for $a(x, y) = A + B(x+y) + Cxy$ $M_1(t) \equiv 1$, $M_2 = M_2(0)$.

Cases	t_g
$C = 0$	$+\infty$
$C \neq 0$: $A = 0$: $B = 0$	$1/CM_2$
: $B \neq 0$	$\frac{1}{2B} \ln \frac{2B + CM_2}{CM_2}$
: $A \neq 0$: $B^2 = AC$	$A/B(A + BM_2)$
: $B^2 > AC$ $\mu_\pm = \frac{-B \pm (B^2 - AC)^{1/2}}{A}$	$\frac{1}{A(\mu_+ - \mu_-)} \ln \frac{\mu_+}{\mu_-} \frac{1-\mu_+ M_2}{1 - \mu_- M_2}$
: $B^2 < AC$ $\alpha = -\frac{B}{A}$, $\gamma = \frac{(AC - B^2)^{1/2}}{A}$	$\frac{1}{A\gamma}\left(\tan^{-1}\frac{\alpha}{\gamma} + \tan^{-1}\frac{1 - \alpha M_2}{\gamma M_2}\right)$

2. Solutions for $t < t_g$

Let us introduce $f_k(x, t)$, $k = 1, 2, 3, \ldots$, functions satisfying

$$\frac{\partial f_k(x,t)}{\partial t} = \frac{1}{2}\sum_{i=1}^{k-1} \int_0^x f_i(y,t) f_{k-i}(x-y,t) a(y, x-y)\, dy - f_k(x,t) \int_0^\infty f(y,t) a(x,y)\, dy$$

$$f_1(x,0) = g(x) \qquad f_2(x,0) = f_3(x,0) = \ldots = 0. \tag{9}$$

Summing over $k = 1, 2, 3, \ldots$, shows that $\sum_{k=1}^\infty f_k(x,t)$ satisfies (1). Hence

$$f(x,t) = \sum_{k=1}^\infty f_k(x,t). \tag{10}$$

$f_k(x,t)$ has a simple physical interpretation: $f_k(x,t)\,dx$ is the volume-average number of particles at time t which have mass x to $x + dx$ *and* which contain k of the initial particles. Equation (9) has a physical interpretation similar to that of equation (1).

Let us parametrise the time by μ, so that

$$\frac{\partial f_k(x)}{\partial \mu}=\frac{-1}{D(\mu)}\sum_{i=1}^{k-1}\int_0^x f_i(y)f_{k-i}(x-y)a(y,x-y)\,dy$$

$$+\frac{2}{D(\mu)}f_k(x)[(A+Bx)\mu+(B+Cx)]. \tag{11}$$

Assume that

$$(k-1)f_k(x)=\frac{1-\mu}{D(\mu)}\sum_{i=1}^{k-1}\int_0^x f_i(y)f_{k-i}(x-y)a(y,x-y)\,dy \qquad k=2,3,\ldots \tag{12}$$

so that

$$\partial f_k(x)/\partial\mu=f_k(x)\{-(k-1)/(1-\mu)+(2/D(\mu))[(A+Bx)\mu+(B+Cx)]\}. \tag{13}$$

Use of table 1 to solve this separable differential equation yields

$$f_k(x,t)=C_k(x)(1-\mu)^{k-1}\exp[-\tfrac{1}{2}(Cx+2B)t+x\mu] \qquad A=0 \tag{14a}$$

$$=C_k(x)(1-\mu)^{k-1}[D(\mu)/D(1)]^{1+(B/A)x}\exp\{-[(AC-B^2)/A]xt\} \qquad A\neq 0 \tag{14b}$$

where μ and t are related by table 1. $C_k(x)$ is determined by the restriction (12) on $f_k(x)$:

$$(k-1)C_k(x)=\sum_{i=1}^{k-1}\int_0^x C_i(y)C_{k-i}(x-y)\frac{a(y,x-y)}{D(1)}\,dy. \tag{15a}$$

Equations (14) and the initial conditions in (9) imply

$$C_1(x)=g(x). \tag{15b}$$

The recursion (15) forces $f_k(x,t)$ in (14) to satisfy both (12) and (13) so that $f(x,t)$ in (10) must be a solution of (1).

Spouge (1983a) showed that assumption (12) leads to solutions of equation (4) only when the kernel has the form (2); this shows the same is true for the more general equation (1).

Branching process models of aggregation prove the most natural derivation of the assumption (12); Spouge (1983b) explores their relation to solutions of equation (1).

Acknowledgments

This work was supported by a Fellowship of the Medical Research Council of Canada.

References

Barrow J D 1981 *J. Phys. A: Math. Gen.* **14** 729

Cohen R J and Benedek G B 1982 *J. Phys. Chem.* **86** 3696

Drake R L 1972 *Topics in current aerosol research, International reviews in aerosol physics and chemistry vol 2* ed G M Hidy and J R Brock (Oxford: Pergamon)

Ernst M H, Hendriks E M and Ziff R M 1982 *J. Phys. A: Math. Gen.* **15** L743
Leyvraz F and Tschudi H R 1981 *J. Phys. A: Math. Gen.* **13** 3389
Lushnikov A A 1973 *J. Coll. Interface Sci.* **45** 549
Popel A S, Regirer S A and Shadrina N Kh 1975 *Prikl. Math. Mech.* **39** 130
Ramabhadran T W, Poeterson T W and Seinfeld J H 1976 *AI Chem. Eng. J.* **22** 840
Spouge J L 1983a *J. Phys. A: Math. Gen.* **16** 767
—— 1983b *Adv. Appl. Prob.* submitted
Ziff R M 1980 *J. Stat. Phys.* **23** 241
Ziff R M, Hendriks E M and Ernst M H 1982 *Phys. Rev. Lett.* **49** 593
Ziff R M and Stell G 1980 *J. Chem. Phys.* **73** 3492