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Gelation time in the discrete coagulation–fragmentation equations with a bilinear coagulation kernel

Éric Brunelle[1], **Robert G Owens**[1] **and Henry J van Roessel**[2]

[1] Département de mathématiques et de statistique, Université de Montréal, CP 6128 succ. Centre-Ville, Montréal, Québec H3C 3J7, Canada

[2] Department of Mathematical and Statistical Sciences, University of Alberta, Edmonton, Alberta T6G 2G1, Canada

E-mail: owens@dms.umontreal.ca

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Abstract

The discrete coagulation–fragmentation equations with a bilinear coagulation kernel, describing the evolution of clusters of particles undergoing binary reactions, are studied. We compute the gelation time and post-gelation mass for the pure coagulation equation. A method of characteristics is developed to solve numerically the partial differential equation satisfied by a moment generating function for a product coagulation kernel and a constant fragmentation rate. The accuracy of the numerical method is verified by comparison of the numerical results with an exact solution for the number density of monomers. For a given coagulation rate, a critical value of the fragmentation rate, values greater than which lead to mass conservation, is identified.

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1. Introduction

The evolution in time of a system of particle clusters, under the assumption of only binary interactions, may be modelled by the discrete coagulation–fragmentation equations

$$\frac{\mathrm{d}N_k}{\mathrm{d}t} = \frac{1}{2}\sum_{i=1}^{k-1} K_{i,k-i} N_i N_{k-i} + \sum_{j=1}^{\infty} F_{k,j} N_{k+j} - \frac{1}{2}\sum_{i=1}^{k-1} F_{i,k-i} N_k - \sum_{j=1}^{\infty} K_{k,j} N_k N_j, \tag{1}$$

where N_k denotes the number density of clusters of k particles (so-called k-mers), $K_{k,j}$ is the coagulation rate of a k-mer with a j-mer (to form a $(k + j)$-mer) and $F_{k,j}$ denotes the rate at which a $(k + j)$-mer fragments into a k-mer and a j-mer. Useful reviews of the numerous application areas of (1) may be found, for example, in chapters by Drake [8] and Ernst [11].

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There exists a large and growing literature discussing the existence and uniqueness of solutions of (1) and its continuous analogue, in the special case of pure coagulation ($F_{k,j} = 0, \forall\, k, j$) and for various choices of coagulation coefficient $K_{k,j}$. A summary of some of the more important papers for the coagulation equation may be found in the new review by Wattis [24], for example.

Existence and uniqueness results for the full discrete coagulation–fragmentation equations are rather less numerous. In 1984, Spouge [22] proved the existence of solutions over some non-zero time interval to (1) in the case $0 \leqslant K_{i,j} \leqslant c_i c_j$, where $c_n = o(n)$ as $n \to \infty$, $\sum_{i=1}^{k-1} \mathrm{i} F_{i,k-i} \leqslant Ck$ for some constant C and $0 \leqslant F_{k,i} \leqslant d_{k,i} = o(k)$ for fixed i as $k \to \infty$. Existence was proved by Spouge by taking the limit of the finite-dimensional system, obtained by replacing ∞ in the upper limits of sums in (1) by $N - k$ for some finite N. A solution of (1) obtained in this way is called an *admissible* solution. A similar approach was adopted by Ball and Carr [1] in their extension to the fragmentation–coagulation equations of earlier analysis of uniqueness and existence of solutions of the Becker–Döring equations [2]. Amongst other results, the authors showed that a unique solution to (1) existed provided $K_{j,k} \leqslant C(jk)^\alpha\, \forall\, j, k$ where C is a positive constant and $0 \leqslant \alpha \leqslant \frac{1}{2}$, and a suitably restrictive growth condition was imposed on the fragmentation rate. Questions of the existence and uniqueness of solutions to (1) are closely related to (and therefore often considered in the same papers as) the issue of mass conservation, that is, whether or not the total particle number density $\sum_{k=0}^{\infty} k N_k$ remains constant for all time. Certain choices of the coagulation and fragmentation rate functions or of the initial conditions may be shown to lead to *gelation*. That is mass conservation breaks down in finite time and is physically interpreted as being caused by the appearance of a 'superparticle' (a cluster of infinite size) that takes mass out of the system (see Ernst *et al* [12]). Ball and Carr [2] were able to show that solutions to (1) having $K_{j,k} \leqslant r_j + r_k + \alpha_{j,k}$ where $\{r_j\}$ is a non-negative sequence, $\alpha_{j,k} \geqslant 0\, \forall\, j, k$ and $\alpha_{j,k} \leqslant C(j+k)$ for C a constant and j, k sufficiently large, were mass conserving, provided there exists a constant κ such that

$$\sum_{j=1}^{[(r+1)/2]} j F_{r-j,j} \leqslant \kappa r, \qquad \forall\, r \geqslant 2,$$

where $[\cdot]$ denotes the integer part of a real number. A couple of years later, Carr [4] considered the case $K_{j,k} \leqslant C(j+k)$ and *strong fragmentation*: there exists $\gamma > 0$ such that for any $m \geqslant 0\, \exists \kappa(m) > 0$ such that

$$\sum_{j=1}^{[(r-1)/2]} j^m F_{r-j,j} \geqslant \kappa(m) r^{\gamma+m}, \qquad \forall\, r \geqslant 3. \tag{2}$$

It was shown in [4] that under the above conditions admissible solutions N_k had finite moments

$$M_n = \sum_{k=1}^{\infty} k^n N_k(t), \tag{3}$$

for any $n > 1$. Carr also showed convergence from arbitrary initial data with finite density to the equilibrium solution. The existence and uniqueness of (mass conserving) admissible solutions for coagulation coefficients satisfying

$$K_{k,j} \leqslant C(jk)^\alpha,$$

for some constant $C, \alpha \in \left(\frac{1}{2}, 1\right)$ and strong fragmentation in the sense of (2) was proved by Da Costa [7] in 1995. More recently, Fournier and Mischler [15] considered the discrete coagulation–fragmentation system (1) with

$$\begin{aligned} &K_{i,j} \leqslant K_0(ij)^\alpha, \qquad F_1(i+j)^s \geqslant F_{i,j} \geqslant F_0(i+j)^\gamma, \\ &\alpha \in [0,1], \quad \gamma \in (-1,\infty), \quad s \in (-1,\infty). \end{aligned} \tag{4}$$

With fragmentation sufficiently strong relative to the coagulation rate in the sense that $2+\gamma > 2\alpha$, and for sufficiently small initial data, the authors were able to prove the exponential convergence of M_2 to a unique equilibrium value.

A number of results are available for the continuous coagulation–fragmentation equations. In this case the coagulation and fragmentation coefficients are continuous functions of variables $(x, y) \in [0,\infty)^2$. Dubovskiĭ and Stewart [9] proved the existence, uniqueness and mass conservation theorems for the continuous analogue to (1) in the case $K(x, y) \leqslant C(1+x+y)$ and a large class of fragmentation kernels F. Escobedo *et al* [13] proved that if

$$K(x,y) = x^\alpha y^\beta + x^\beta y^\alpha, \qquad F(x,y) = (1+x+y)^\gamma,$$

with $0 \leqslant \alpha \leqslant \beta \leqslant 1$ and $\gamma \in \mathbb{R}$, gelation occurred if $\lambda := \alpha+\beta > 1$ and $\gamma < (\lambda-3)/2$. Shortly afterwards, Escobedo *et al* [14] showed that for $\lambda > 1$ mass was conserved when $\gamma > \lambda - 2$ (that is, for sufficiently strong fragmentation) but that for sufficiently large initial data gelation occurred when $\lambda > 1$ and $\gamma \in [(\lambda-3)/2, \lambda-2)$.

A common theme running through many of the papers cited above, and others like them, is the identification, for a given coagulation rate, of what constitutes a sufficiently strong fragmentation rate in order that mass conserving solutions should exist. In the present paper, we wish to study the behaviour of solutions to (1) when the coagulation kernel has the bilinear form

$$K_{k,j} = (\alpha+\beta k)(\alpha+\beta j) = \alpha^2 + \alpha\beta(k+j) + \beta^2 kj,$$

for real constants α and β and the fragmentation kernel assumes its simplest possible form $F_{k,j} = b$, where b is a constant. It is known that for the pure coagulation equation ($b = 0$), in either its discrete or continuous form, gelation occurs for any $\beta > 0$ [17–21]. By choosing $b > 0$ it is to be expected that the gelation time will increase. In this paper, we wish to investigate whether by choosing b sufficiently large we are able to not just delay the onset of gelation but suppress gelation altogether. If so, how large should b be relative to β in order that the solution be mass conserving and what is the relationship between the gelation time and b?

In section 2 we derive the partial differential equation satisfied by a moment generating function φ and discuss the macroscopic rate equations. We begin section 3 by solving the characteristic equations for the φ equation and deriving expressions for the gelation time and post-gelation mass in the case of the pure coagulation equation. The remainder of the paper is concerned, when $\alpha = 0$, with determining how large b should be relative to β in order that the mass should be conserved. We begin in section 3.2.1 by showing, using elementary arguments, that a critical value of $\varepsilon := b/(\beta^2 M_1) \in [1, 6]$ must exist, values greater than or equal to which ensure that the solution to (1) is mass conserving. The theory for regular perturbation solutions for small values of ε is derived in section 3.2.2. In section 3.2.3, we present a numerical method for the integration of the characteristic equations referred to above and use our method to determine numerically the gelation time as a function of ε.

2. Basic equations

2.1. Moment generating function

Let us introduce a moment generating function

$$\varphi(x,t)=\sum_{k=1}^{\infty}x^kN_k(t),\qquad |x|\leqslant 1, \tag{5}$$

and suppose that the first moment (total number of particles) is finite, i.e.

$$M_1=\sum_{k=1}^{\infty}kN_k<\infty.$$

Then, multiplying (1) throughout by x^k and summing over all k we get a partial differential equation for φ

$$\begin{aligned}\frac{\partial\varphi}{\partial t}&=\frac{1}{2}\sum_{k=1}^{\infty}\sum_{i=1}^{k-1}x^kK_{i,k-i}N_iN_{k-i}+\sum_{k=1}^{\infty}\sum_{j=1}^{\infty}x^kF_{k,j}N_{k+j}\\&\quad-\frac{1}{2}\sum_{k=1}^{\infty}\sum_{i=1}^{k-1}x^kF_{i,k-i}N_k-\sum_{k=1}^{\infty}\sum_{j=1}^{\infty}x^kK_{k,j}N_kN_j, &(6)\\&=\frac{1}{2}\sum_{k=1}^{\infty}\sum_{j=1}^{\infty}x^{k+j}K_{k,j}N_kN_j+\sum_{k=1}^{\infty}\sum_{i=1}^{k-1}x^iF_{i,k-i}N_k\\&\quad-\frac{1}{2}\sum_{k=1}^{\infty}\sum_{i=1}^{k-1}x^kF_{i,k-i}N_k-\sum_{k=1}^{\infty}\sum_{j=1}^{\infty}x^kK_{k,j}N_kN_j, &(7)\\&=\frac{1}{2}\sum_{k=1}^{\infty}\sum_{j=1}^{\infty}x^{k+j}(\alpha^2+\alpha\beta(k+j)+\beta^2kj)N_kN_j\\&\quad+b\sum_{k=1}^{\infty}\left(\frac{x^k-x}{x-1}\right)N_k-\frac{b}{2}\sum_{k=1}^{\infty}x^k(k-1)N_k\\&\quad-\sum_{k=1}^{\infty}\sum_{j=1}^{\infty}x^k(\alpha^2+\alpha\beta(k+j)+\beta^2kj)N_kN_j,\\\Rightarrow\frac{\partial\varphi}{\partial t}&=\frac{\alpha^2\varphi^2}{2}-\alpha^2M_0\varphi+\alpha\beta\left(x\varphi\frac{\partial\varphi}{\partial x}-M_0x\frac{\partial\varphi}{\partial x}-M_1\varphi\right)\\&\quad+\beta^2\left(\frac{1}{2}x^2\left(\frac{\partial\varphi}{\partial x}\right)^2-M_1x\frac{\partial\varphi}{\partial x}\right)+\frac{b}{x-1}(\varphi-xM_0)+\frac{b}{2}\varphi-\frac{bx}{2}\frac{\partial\varphi}{\partial x}. &(8)\end{aligned}$$

In the developments leading to (8) we have used the fact that $M_1<\infty$ means that the series

$$\sum_{k=1}^{\infty}\sum_{j=1}^{\infty}x^{k+j}K_{k,j}N_kN_j,\qquad \sum_{k=1}^{\infty}\sum_{i=1}^{k-1}x^iF_{i,k-i}N_k=b\sum_{k=1}^{\infty}\left(\frac{x^k-x}{x-1}\right)N_k$$

are both (absolutely) convergent for $|x|\leqslant 1$. This allows us to equate the first two terms in (6) with the first two terms in (7), since the first (respective, second) term in (6) is just a rearrangement of the first (respective, second) term in (7).

2.2. Macroscopic rate equations

The evolution equation for the kth moment M_k, assuming that $M_{k+1} < \infty$ is (see, for example, equation (2.2) of [15] or equation (1.6) of [22])

$$\frac{\mathrm{d}M_k}{\mathrm{d}t} = \frac{1}{2}\sum_{i=1}^{\infty}\sum_{j=1}^{\infty} N_i N_j[(i+j)^k - i^k - j^k]K_{ij} + \frac{1}{2}\sum_{i=1}^{\infty} N_i \sum_{j=1}^{i-1}[j^k + (i-j)^k - i^k]F_{j,i-j}. \tag{9}$$

When $k = 1$ and M_2 is finite, we therefore see from (9) that

$$\frac{\mathrm{d}M_1}{\mathrm{d}t} = 0,$$

so that mass is conserved. To see what happens to $\mathrm{d}M_1/\mathrm{d}t$ in the more general case, we differentiate (8) throughout with respect to x and after some tedious but straightforward algebra we get

$$\begin{aligned}\frac{\partial^2\varphi}{\partial t\partial x} &= \left(\alpha(\varphi - M_0) + \beta\left(x\frac{\partial\varphi}{\partial x} - M_1\right)\right)\left((\alpha+\beta)\frac{\partial\varphi}{\partial x} + \beta x\frac{\partial^2\varphi}{\partial x^2}\right) \\ &\quad + b\left(\frac{(x-1)\left(\frac{\partial\varphi}{\partial x} - M_0\right) - (\varphi - xM_0)}{(x-1)^2} - \frac{x}{2}\frac{\partial^2\varphi}{\partial x^2}\right).\end{aligned} \tag{10}$$

Thus,

$$\frac{\mathrm{d}M_1}{\mathrm{d}t} = \lim_{x\to 1^-}\left\{\frac{b}{2}(1-x) + \beta x\left[\alpha(\varphi - M_0) + \beta\left(x\frac{\partial\varphi}{\partial x} - M_1\right)\right]\right\}\frac{\partial^2\varphi}{\partial x^2}. \tag{11}$$

If M_2 is finite then the mass is conserved as before. If, however, $\partial^2\varphi/\partial x^2$ diverges as $x \to 1^-$ then gelation occurs.

In the case $k = 0$, (9) becomes

$$\frac{\mathrm{d}M_0}{\mathrm{d}t} = -\frac{1}{2}(\alpha M_0 + \beta M_1)^2 + \frac{1}{2}b(M_1 - M_0). \tag{12}$$

This is just a non-homogeneous Bernoulli differential equation and if we assume mass conservation then its solution is

$$M_0 - M_{0,\mathrm{st}} = \frac{(M_0(0) - M_{0,\mathrm{st}})}{\exp(\gamma t/2) + \frac{\alpha^2}{\gamma}(\exp(\gamma t/2) - 1)(M_0(0) - M_{0,\mathrm{st}})}, \tag{13}$$

where

$$\gamma = \sqrt{b^2 + 4\alpha b M_1(\alpha+\beta)},$$

$M_0(0)$ denotes the initial value of M_0 and $M_{0,\mathrm{st}}$ is the steady value of M_0, given by

$$M_{0,\mathrm{st}} = \begin{cases} \dfrac{\gamma - b}{2\alpha^2} - \dfrac{\beta M_1}{\alpha}, & \alpha \neq 0 \\ M_1\left(1 - \dfrac{\beta^2 M_1}{b}\right), & \alpha = 0. \end{cases} \tag{14}$$

2.3. Characteristic equations for (8)

Let us introduce the variables

$$p := \frac{\partial \varphi}{\partial x}, \qquad q := \frac{\partial \varphi}{\partial t}, \qquad z := \varphi.$$

Then, equation (8) for φ may be written as $F(x, t, z, p, q) = 0$ where $F(x, t, z, p, q)$ is defined as

$$F(x, t, z, p, q) := q - \frac{\alpha^2 z^2}{2} + \alpha^2 M_0 z - \alpha\beta(x(z - M_0)p - M_1 z)$$
$$- \beta^2 \left(\frac{1}{2}x^2 p^2 - x M_1 p\right) - b\left[\frac{1}{2}(z - xp) + \frac{z - xM_0}{x - 1}\right]. \tag{15}$$

In terms of the characteristic variables s and ξ, Charpit's equations become

$$\frac{\mathrm{d}t}{\mathrm{d}s} = \frac{\partial F}{\partial q} = 1, \qquad t(0) = t_0(\xi), \tag{16}$$

$$\frac{\mathrm{d}x}{\mathrm{d}s} = \frac{\partial F}{\partial p} = -\alpha\beta x(z - M_0) - \beta^2(x^2 p - xM_1) + \frac{bx}{2}, \qquad x(0) = x_0(\xi), \tag{17}$$

$$\frac{\mathrm{d}z}{\mathrm{d}s} = p\frac{\partial F}{\partial p} + q\frac{\partial F}{\partial q} = -\alpha\beta xp(z - M_0) - \beta^2(x^2p^2 - xpM_1) + \frac{bxp}{2} + q,$$
$$z(0) = z_0(\xi), \tag{18}$$

$$\frac{\mathrm{d}p}{\mathrm{d}s} = -\frac{\partial F}{\partial x} - p\frac{\partial F}{\partial z} = \alpha^2 p(z - M_0) + \alpha\beta p(z - M_0 + xp - M_1)$$
$$+ \beta^2 p(xp - M_1) + b\left[\frac{p}{x - 1} - \frac{z - M_0}{(x - 1)^2}\right], \qquad p(0) = p_0(\xi), \tag{19}$$

$$\frac{\mathrm{d}q}{\mathrm{d}s} = -\frac{\partial F}{\partial t} - q\frac{\partial F}{\partial z} = \alpha^2(q(z - M_0) - z\dot{M}_0) + \alpha\beta(q(xp - M_1) - xp\dot{M}_0 - z\dot{M}_1)$$
$$- \beta^2 xp\dot{M}_1 + b\left[\frac{q}{2} + \frac{q - x\dot{M}_0}{x - 1}\right], \qquad q(0) = q_0(\xi). \tag{20}$$

We set the initial conditions for t, x and z as

$$t_0(\xi) = 0, \qquad x_0(\xi) = \xi, \qquad \text{and} \qquad z_0(\xi) = h(\xi), \tag{21}$$

for some function $h \in C^\infty[0, 1]$. It then follows from

$$z_0'(\xi) = p_0(\xi)x_0'(\xi) + q_0(\xi)t_0'(\xi)$$

and

$$F(x_0(\xi), t_0(\xi), z_0(\xi), p_0(\xi), q_0(\xi)) = 0$$

that

$$p_0(\xi) = h'(\xi) \tag{22}$$

and

$$q_0(\xi) = \frac{\alpha^2 h^2(\xi)}{2} - \alpha^2 M_0(0)h(\xi) + \alpha\beta[\xi(h(\xi) - M_0(0))h'(\xi) - M_1(0)h(\xi)]$$
$$+ \beta^2\left[\frac{1}{2}\xi^2 h'(\xi)^2 - \xi M_1(0)h'(\xi)\right] + b\left[\frac{1}{2}(h(\xi) - \xi h'(\xi)) + \frac{h(\xi) - \xi M_0(0)}{\xi - 1}\right]. \tag{23}$$

We observe that

$$M_0(0) = \varphi(1,0) = z_0(1) = h(1),$$

and

$$M_1(0) = p_0(1) = h'(1)$$

so that (23) becomes

$$q_0(\xi) = \frac{\alpha^2 h^2(\xi)}{2} - \alpha^2 h_1 h(\xi) + \alpha\beta[\xi(h(\xi) - h_1)h'(\xi) - h_1' h(\xi)] + \beta^2\left[\frac{1}{2}\xi^2 h'(\xi)^2 - \xi h_1' h'(\xi)\right] + b\left[\frac{1}{2}(h(\xi) - \xi h'(\xi)) + \frac{h(\xi) - \xi h_1}{\xi - 1}\right], \quad (24)$$

where, in the above, we have introduced the abbreviated notation h_1 and h_1' for $h(1)$ and $h'(1)$, respectively.

Clearly, $t = s$ and we now seek solutions to (16)–(24) in the form $x = X(\xi, t), z = Z(\xi, t), p = P(\xi, t)$ and $q = Q(\xi, t)$. Provided $\partial X/\partial\xi \neq 0$ we may invert the relationship between x and ξ to get $\xi = \widehat{\xi}(x, t)$ for some function $\widehat{\xi} : [0, 1] \times \mathbb{R} \longrightarrow [0, 1]$ and then write

$$\varphi(x, t) = Z(\widehat{\xi}(x, t), t), \qquad \frac{\partial\varphi}{\partial x}(x, t) = P(\widehat{\xi}(x, t), t).$$

It follows that

$$M_0(t) = \varphi(1, t) = Z(\widehat{\xi}(1, t), t) = Z(\xi_0(t), t),$$
$$M_1(t) = \frac{\partial\varphi}{\partial x}(1, t) = P(\xi_0(t), t) = \frac{\partial Z}{\partial\xi}(\xi_0(t), t)\frac{\partial\widehat{\xi}}{\partial x}(1, t) = \frac{\frac{\partial Z}{\partial\xi}(\xi_0(t), t)}{\frac{\partial X}{\partial\xi}(\xi_0(t), t)},$$

where $\xi_0(t)$ is defined by $X(\xi_0(t), t) = 1$.

3. Solution of (8)

The solution to equation (8) in the case $\beta = 0$ has been known since 1945 for monodisperse initial conditions [3] and the solution for more general initial conditions may be found in [11]. Solutions and gelation times for the pure coagulation equation ($b = 0$) with a general quadratic coagulation kernel $K_{ij} = A + B(i + j) + Cij$ and monodisperse initial conditions have been obtained by Spouge [21] and showed that the gelation time was finite for any $C > 0$. Indeed, for $A = 0$ and $C > 0$ this gelation time was calculated as $t_{\mathrm{gel}} = 1/C$. The case $A = B = 0$ and $C = 1$ with monodisperse initial conditions had already been considered by McLeod [18] and he showed, by explicitly calculating the solution, that a solution to this particular case of the coagulation equations existed for $0 \leqslant t \leqslant 1$.

In the present paper, we present the following results:

(1) We compute the gelation time and post-gelation mass for the coagulation equation when $\alpha = 0$ and the initial conditions are as given in (21). In doing so, we reproduce the formula obtained by Hendriks *et al* [16] for the gelation time in the case of general initial conditions and generalize the post-gelation mass results of McLeod [18], Spouge [21] and Hendriks *et al* [16], obtained under the assumption of monodisperse initial conditions. A similar analysis in the case of the continuous coagulation equations may be found in van Roessel and Shirvani [23].

(2) A method of characteristics is developed to solve (8) numerically for general values of β and b, in the case $\alpha = 0$. A critical value of ε is identified, values smaller than which lead to a finite gelation time.

3.1. Solution of (8) when $\alpha = 0$, $b = 0$, $\beta > 0$. Gelation time and post-gelation mass

The characteristic equations (16)–(24) in this case become

$$\frac{\mathrm{d}x}{\mathrm{d}t} = -\beta^2 x(xp - M_1(t)), \qquad x(0) = \xi, \tag{25}$$

$$\frac{\mathrm{d}z}{\mathrm{d}t} = -\beta^2 xp(xp - M_1(t)) + q, \qquad z(0) = h(\xi), \tag{26}$$

$$\frac{\mathrm{d}p}{\mathrm{d}t} = \beta^2 p(xp - M_1(t)), \qquad p(0) = h'(\xi), \tag{27}$$

$$\frac{\mathrm{d}q}{\mathrm{d}t} = -\beta^2 xp\dot{M}_1(t), q(0) = \frac{\beta^2}{2}\xi h'(\xi)[\xi h'(\xi) - 2h_1']. \tag{28}$$

From (25) and (27) we get

$$\frac{\mathrm{d}p}{\mathrm{d}x} = -\frac{p}{x} \quad \Rightarrow \quad px = \xi h'(\xi). \tag{29}$$

Then, from (28) and (29) it may be deduced that

$$\frac{\mathrm{d}q}{\mathrm{d}t} = -\beta^2 \xi h'(\xi)\dot{M}_1(t) \quad \Rightarrow \quad q = Q(\xi, t) = \frac{\beta^2}{2}\xi h'(\xi)[\xi h'(\xi) - 2M_1(t)]. \tag{30}$$

(In fact, in the present case, we could have got q directly from (15).) Equations (25) and (29) lead to

$$\frac{\mathrm{d}x}{\mathrm{d}t} = -\beta^2 x(\xi h'(\xi) - M_1(t)) \quad \Rightarrow \quad x = X(\xi, t) = \xi \exp(-\beta^2[t\xi h'(\xi) - R(t)]), \tag{31}$$

where

$$R(t) := \int_0^t M_1(s)\,\mathrm{d}s,$$

and from (29) and (31) we see that

$$p = P(\xi, t) = h'(\xi)\exp(\beta^2[t\xi h'(\xi) - R(t)]).$$

Finally, we use (26), (29) and (30) in order to conclude that

$$\frac{\mathrm{d}z}{\mathrm{d}t} = -\frac{\beta^2\xi^2 h'(\xi)^2}{2} \quad \Rightarrow \quad z = Z(\xi, t) = h(\xi) - \frac{\beta^2 t\xi^2 h'(\xi)^2}{2}.$$

Gelation will occur when there is a shock in the solution of (8) and this occurs when a derivative of $\partial\varphi/\partial x$ becomes infinite or, equivalently, when $\partial X/\partial\xi = 0$.

Differentiating X from (31) we get

$$\frac{\partial X}{\partial \xi}(\xi, t) = \exp(-\beta^2[t\xi h'(\xi) - R(t)])[1 - \beta^2 t\xi(\xi h''(\xi) + h'(\xi))],$$

so that

$$\frac{\partial X}{\partial \xi} = 0 \quad \Rightarrow \quad t = \frac{1}{\beta^2\xi(\xi h''(\xi) + h'(\xi))} =: T(\xi).$$

Defining $T(\xi)$ as in the above equation, the gelation time t_{gel} is now given by

$$t_{\mathrm{gel}} := \inf_{0\leqslant\xi\leqslant 1} T(\xi), \tag{32}$$

and since both h' and h'' must be monotonic increasing functions, T is a monotonic decreasing function, so that

$$t_{\mathrm{gel}} = T(1) = \frac{1}{\beta^2(h_1'' + h_1')} = \frac{1}{\beta^2 M_2(0)}. \tag{33}$$

When $\beta = 1$ this is the same result as that of Hendricks *et al* [16] (see their equation (5.7)). When monodisperse initial conditions are imposed ($h(\xi) = M_1(0)\xi$) we see that the gelation time t_{gel} equals $1/(\beta^2 M_1(0))$ which is the same result (after a change of notation) obtained by Spouge [21].

We now differentiate $X(\xi_0(t), t) = 1$ throughout with respect to t to get

$$\frac{\partial X}{\partial \xi}(\xi_0(t), t)\frac{\mathrm{d}\xi_0}{\mathrm{d}t} + \frac{\partial X}{\partial t}(\xi_0(t), t) = 0. \tag{34}$$

From (25) we see that when $X = 1$

$$\frac{\partial X}{\partial t} = \frac{\mathrm{d}x}{\mathrm{d}t} = -\beta^2(P(\xi_0(t), t) - M_1(t)) = 0.$$

Prior to gelation ($0 \leqslant t < t_{\mathrm{gel}}$) we have $\partial X/\partial \xi \neq 0$ so that from (34)

$$\frac{\mathrm{d}\xi_0(t)}{\mathrm{d}t} = 0 \quad \Rightarrow \quad \xi_0(t) = 1.$$

For $t \geqslant t_{\mathrm{gel}}$

$$\frac{\partial X}{\partial \xi}(\xi_0, t) = 0,$$

which means that $\xi = \xi_0(t)$ is the solution to the equation

$$1 - \beta^2 t\xi_0[h'(\xi_0) + \xi_0 h''(\xi_0)] = 0.$$

Once ξ_0 has been determined, the post-gelation mass $M_1(t)$ is calculated using (29) as

$$M_1(t) = P(\xi_0(t), t) = \frac{\xi_0(t)h'(\xi_0(t))}{X(\xi_0(t), t)} = \xi_0(t)h'(\xi_0(t)).$$

This generalizes the result, obtained by previous authors (see, for example, Hendricks *et al* [16] and the references therein) for monodisperse initial conditions, that, for $t \geqslant t_{\mathrm{gel}}$

$$M_1(t) = \frac{M_1(0)}{\beta^2 t}.$$

Remark. In the case of the pure coagulation equations, a product coagulation kernel $K_{ij} = \beta^2 ij$ and under monodisperse initial conditions, the exact solution to (1) is known [8, 17, 18, 24]:

$$N_k(t) = \begin{cases} \beta^{2(k-1)} t^{k-1} \exp(-k\beta^2 t)\dfrac{k^{k-2}}{k!} & t \leqslant \dfrac{1}{\beta^2}, \\ \exp(-k)\dfrac{k^{k-2}}{\beta^2 t k!} & t \geqslant \dfrac{1}{\beta^2}, \end{cases} \tag{35}$$

and allows us to state for this case that all moments M_k remain finite up to t_{gel}. All M_k for $k \geqslant 2$ then diverge at $t = t_{\mathrm{gel}}$. Evidence for the existence of only one singular point t_{gel} in the case of more general coagulation kernels has been presented in section 5.1 of [16], for example. However, the question of whether or not higher moments (M_k, $k \geqslant 3$) diverge before the gel time in the general case, including the full fragmentation–coagulation equations, still appears to be an open one. From (9) it may be seen that the fragmentation terms have a negative net effect on the rate of change of M_k. This is because for all $k \geqslant 1$ and $i \geqslant 1$,

$$\begin{aligned} \sum_{j=1}^{i-1}[j^k + (i-j)^k - i^k] &= 2\sum_{j=1}^{i-1} j^k - i^k(i-1) \leqslant 2\int_{1/2}^{i-1/2} x^k dx - i^k(i-1) \\ &= \frac{2}{k+1}\left[\left(i - \frac{1}{2}\right)^{k+1} - \frac{1}{2^{k+1}}\right] - i^{k+1} + i^k \leqslant 0. \end{aligned} \tag{36}$$

It follows that starting with the same initial conditions, the solution M_k of (9) is bounded above by the corresponding solution to the pure coagulation equation up to the pure coagulation gel time. Thus, (9) certainly remains valid for all k over this time interval. We assume in what follows that all higher moments ($k \geqslant 2$) diverge (if at all) at the same time: the fragmentation–coagulation gel time, which is, of course, greater than the pure coagulation gel time.

3.2. *Solution of (8) when* $\alpha = 0, \beta > 0, b > 0$.

For simplicity we shall set $\alpha = 0$ throughout this section, since choosing $\alpha \neq 0$ does not change the qualitative gelling behaviour of the solutions (in particular, if a solution is mass conserving with $\alpha = 0$ it remains so with $\alpha > 0$ [21]). We rescale each of the $N_k (k = 1, 2, 3, \ldots)$ by dividing them by M_1. We introduce a non-dimensional time $t^* = \beta^2 M_1 t/2$ and denote the ratio $b/(\beta^2 M_1)$ by ε.

3.2.1. Bounds on the gelation time. Dropping the asterisk on the dimensionless time, we may deduce from (9) in the case $k = 2$ that (with the assumption, as noted above, that M_3 remains finite as long as M_2 does)

$$\frac{\mathrm{d}M_2}{\mathrm{d}t} = 2M_2^2 - \frac{\varepsilon}{3}(M_3 - 1). \tag{37}$$

Then, inspired by Escobedo *et al* [13] (see their equations (1.34) and (1.35)), we see that since, by Hölder's inequality, $M_3 \geqslant M_2^2$,

$$\frac{\mathrm{d}M_2}{\mathrm{d}t} \leqslant \left(2 - \frac{\varepsilon}{3}\right) M_2^2 + \frac{\varepsilon}{3}. \tag{38}$$

Therefore,

$$M_2(t) \leqslant \begin{cases} M_2(0) + 2t & \text{for} \quad \varepsilon = 6, \\ \frac{1}{\sqrt{1-6\varepsilon^{-1}}} \left[\frac{(1+\sqrt{1-6\varepsilon^{-1}}M_2(0)) - (1-\sqrt{1-6\varepsilon^{-1}}M_2(0)) \exp\left(-\frac{2\varepsilon t\sqrt{1-6\varepsilon^{-1}}}{3}\right)}{(1+\sqrt{1-6\varepsilon^{-1}}M_2(0)) + (1-\sqrt{1-6\varepsilon^{-1}}M_2(0)) \exp\left(-\frac{2\varepsilon t\sqrt{1-6\varepsilon^{-1}}}{3}\right)} \right] & \text{for} \quad \varepsilon > 6, \end{cases} \tag{39}$$

so that M_2 remains bounded for all finite t and the solutions to (1) are mass conserving, whenever $\varepsilon \geqslant 6$, the right-hand sides of (39) being the exact solutions to (38) when equality replaces the inequality.

When $\varepsilon < 6$

$$M_2(t) \leqslant \frac{1}{\sqrt{6\varepsilon^{-1} - 1}} \tan\left(\frac{\varepsilon t\sqrt{6\varepsilon^{-1} - 1}}{3} + \tan^{-1}(\sqrt{6\varepsilon^{-1} - 1}M_2(0))\right). \tag{40}$$

This does not mean necessarily that the system has a finite gelation time but does imply that the gelation time must exceed

$$t_{\mathrm{gel}}^{\min} := \frac{3}{\varepsilon\sqrt{6\varepsilon^{-1} - 1}} \left[\frac{\pi}{2} - \tan^{-1}(\sqrt{6\varepsilon^{-1} - 1}M_2(0))\right], \tag{41}$$

this being the time at which the bound in (40) becomes infinite. In an attempt to find an upper bound on the gelation time we first write the rescaled form of (12) when $\alpha = 0$ as

$$\frac{\mathrm{d}M_0}{\mathrm{d}t} = (\varepsilon - 1) - \varepsilon M_0, \tag{42}$$

and, under the assumption of mass conservation, solve to get

$$M_0 = (1 - \varepsilon^{-1}) + (M_0(0) - (1 - \varepsilon^{-1})) \exp(-\varepsilon t). \tag{43}$$

On physical grounds we must have $M_0(0) > 0$ and clearly, when $\varepsilon > 1$, M_0 as given above never attains zero. Still supposing that M_1 is a constant, we calculate the (finite) time at which M_0 as given by (43) vanishes when $\varepsilon < 1$ to be $t_{\text{gel}}^{\max}$, where this is

$$t_{\text{gel}}^{\max} = \frac{1}{\varepsilon}\log\left(\frac{\varepsilon(M_0(0)-1)+1}{1-\varepsilon}\right).$$

Since, from Hölder's inequality

$$M_2 \geqslant \frac{1}{M_0},$$

we conclude that when $\varepsilon < 1$ the gelation time is finite and bounded above by $t_{\text{gel}}^{\max}$.

Remark. In the continuous case, corollary 2.5 of [14], for example, shows that when the initial mass $M_1(0)$ is big enough (thus, in our notation, when ε is sufficiently small) gelation takes place.

3.2.2. A perturbation solution to (8). We seek a regular perturbation expansion, supposed to be valid for sufficiently small ε for N_k:

$$N_k = N_{k0} + \varepsilon N_{k1} + \varepsilon^2 N_{k2} + \cdots, \qquad k = 1, 2, 3, \ldots,$$

which leads to expansions for the moments and moment generating function of the form

$$\begin{aligned} M_k &= M_{k0} + \varepsilon M_{k1} + \varepsilon^2 M_{k2} + \cdots, \qquad k = 0, 1, 2, \ldots, \\ \varphi &= \varphi_0 + \varepsilon\varphi_1 + \varepsilon^2\varphi_2 + \cdots. \end{aligned} \tag{44}$$

Substituting the perturbation expansions (44) into the rescaled form of (8)

$$\frac{\partial\varphi}{\partial t} = x^2\left(\frac{\partial\varphi}{\partial x}\right)^2 - 2x\frac{\partial\varphi}{\partial x} + \frac{2\varepsilon}{x-1}(\varphi - xM_0) + \varepsilon\left(\varphi - x\frac{\partial\varphi}{\partial x}\right), \tag{45}$$

leads to

$$\begin{aligned} &\frac{\partial\varphi_0}{\partial t} + \varepsilon\frac{\partial\varphi_1}{\partial t} + \varepsilon^2\frac{\partial\varphi_2}{\partial t} + \cdots \\ &\quad = x^2\left(\frac{\partial\varphi_0}{\partial x} + \varepsilon\frac{\partial\varphi_1}{\partial x} + \varepsilon^2\frac{\partial\varphi_2}{\partial x} + \cdots\right)^2 - 2x\left(\frac{\partial\varphi_0}{\partial x} + \varepsilon\frac{\partial\varphi_1}{\partial x} + \varepsilon^2\frac{\partial\varphi_2}{\partial x} + \cdots\right) \\ &\quad + \frac{2\varepsilon}{(x-1)}(\varphi_0 + \varepsilon\varphi_1 + \varepsilon^2\varphi_2 + \cdots - x(M_{00} + \varepsilon M_{01} + \varepsilon^2 M_{02} + \cdots)) \\ &\quad + \varepsilon\left(\varphi_0 + \varepsilon\varphi_1 + \varepsilon^2\varphi_2 + \cdots - x\left(\frac{\partial\varphi_0}{\partial x} + \varepsilon\frac{\partial\varphi_1}{\partial x} + \varepsilon^2\frac{\partial\varphi_2}{\partial x} + \cdots\right)\right). \end{aligned} \tag{46}$$

We seek to solve (45) subject to the usual initial conditions $x(0) = \xi$ and $\varphi(x(0), 0) = h(\xi)$. The $O(1)$ solution to (45) is just that of the pure coagulation case ($b = 0$) and the method of solution therefore follows directly from that used in section 3.1. From Charpit's equations we solve to get

$$x(t) = \xi\exp(2t(1 - \xi h'(\xi))), \tag{47}$$

$$\varphi_0(x(t), t) = h(\xi) - t\xi^2 h'(\xi)^2. \tag{48}$$

Under monodisperse initial conditions the exact solution φ_0 for $0 \leqslant t \leqslant 1/2$ is known (see equation (35)):

$$\varphi_0(x, t) = \sum_{k=1}^{\infty} 2^{k-1} t^{k-1}\exp(-2kt)\frac{k^{k-2}}{k!}x^k. \tag{49}$$

Solving (45) at $O(\varepsilon^k)(k = 1, 2, 3, \ldots)$ necessitates the solution of

$$\frac{\partial \varphi_k}{\partial t} - 2x \frac{\partial \varphi_k}{\partial x}\left(x \frac{\partial \varphi_0}{\partial x} - 1 \right) = F_k(\varphi_0, \varphi_1, \ldots, \varphi_{k-1}), \tag{50}$$

for some function F_k of the (already known) lower order terms in the perturbation expansion. The method of characteristics for (50) would then lead to the following system of equations:

$$\frac{\mathrm{d}x}{\mathrm{d}t} = -2x\left(x \frac{\partial \varphi_0}{\partial x} - 1 \right), \qquad x(0) = \xi, \tag{51}$$

$$\frac{\mathrm{d}z}{\mathrm{d}t} = F_k(\varphi_0(x(t), t), \varphi_1(x(t), t), \ldots, \varphi_{k-1}(x(t), t)),\ z(0) = 0. \tag{52}$$

Although a numerical method could be used to solve (51) and (52) for $k = 1, 2, 3, \ldots$ it is more efficient to solve (45) directly using the method of characteristics. We describe this method in the following section.

3.2.3. A numerical method for the solution of (45). The usual approach to the numerical solution of the discrete coagulation–fragmentation equations for various choices of $K_{k,j}$ and $F_{k,j}$ has been to solve the system (1) approximately by replacing the upper limit ∞ on the second and fourth sums with some large but finite value [5, 6, 10]. The large nonlinear coupled system may then be integrated numerically using Euler's method [6] or a Runge–Kutta method [10], for example. In the present paper, we propose a numerical method for the solution of the partial differential equation (45), satisfied by the moment generating function. For simplicity, we consider the case of monodisperse initial conditions but nothing would prevent us from performing calculations with more general initial data.

When $\alpha = 0$, with monodisperse initial conditions and assuming mass conservation, Charpit's equations (16)–(20) may be written in non-dimensional form

$$\frac{\mathrm{d}x}{\mathrm{d}t} = (\varepsilon + 2)x - 2x^2 p, \qquad x(0) = \xi, \tag{53}$$

$$\frac{\mathrm{d}z}{\mathrm{d}t} = -x^2 p^2 + \varepsilon \left(\frac{x+1}{x-1} \right) z - \frac{2\varepsilon x M_0}{(x-1)}, \qquad z(0) = \xi, \tag{54}$$

$$\frac{\mathrm{d}p}{\mathrm{d}t} = 2p(xp-1) + 2\varepsilon \left(\frac{p}{(x-1)} - \frac{(z - M_0)}{(x-1)^2} \right), \qquad p(0) = 1. \tag{55}$$

We could, of course, have written the system as a coupled set of four equations (for x, z, p and q), but since we have equation (45), relating $\partial\varphi/\partial t$ to φ, $\partial\varphi/\partial x$ and M_0, this is not necessary.

The numerical method proposed in this paper assumes mass conservation (M_1 a constant) and therefore the exact expression for M_0 from (43). This means, in particular, that the numerical solutions obtained will only be valid up to the gelation time. However, since the purpose of our numerical algorithm is to determine the gelation time as a function of ε, this is not a handicap. The initial mesh spacing is chosen as $\Delta x := 1/N$ for some $N \in \mathbb{Z}^+$, and at $t = 0$ the mesh points are $x_1, \ldots, x_{N-1}$ with $x_i = \mathrm{i}\Delta x$. A time step Δt is set and the numerical solutions to equations (53)–(55) advanced in time using a second-order Runge–Kutta method. The number of time steps (m, say) for which this is done is limited by the constraint that the characteristic whose foot is at x_{N-1} must not cross $x = 1$. If taking one more time step of size Δt would result in committing such a transgression, the time marching is halted and the solution φ known at all points $X(x_i, m\Delta t)$ $(i = 1, \ldots, N-1)$ is interpolated back onto a

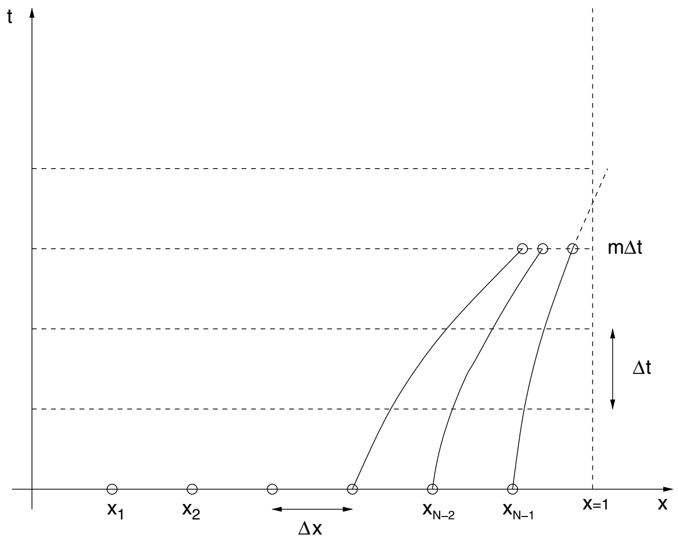

Figure 1. Diagrammatic representation of the proposed method of characteristics for (45).

uniform grid, identical to that at $t = 0$. M_1 may be calculated using a simple backward finite difference, based on the values of φ at $x = 1$ (which is just M_0 and known from (43)) and at $X(x_{N-1}, m\Delta t)$. The process is now repeated. Figure 1 illustrates the method.

3.2.4. Numerical results. By differentiating φ k times with respect to x and setting $x = 0$ we see that we may express the number density of k-mers in the form

$$N_k = \frac{1}{k!} \left. \frac{\partial^k \varphi}{\partial x^k} \right|_{x=0}. \tag{56}$$

The first test of our numerical method, then, will be to compare the result of computing N_k from (1) with that coming from the evaluation of the right-hand side of (56). When $k = 1$ and $\alpha = 0$, for example, the solution to the non-dimensionalized form of (1) up to the gelation time is

$$N_1(\varepsilon, t) = \exp(-2(1+\varepsilon)t) - \left(\frac{1-\varepsilon}{1+\varepsilon}\right)(1 - \exp(-2(1+\varepsilon)t)) + \frac{2}{(2+\varepsilon)}(\exp(-\varepsilon t) - \exp(-2(1+\varepsilon)t)). \tag{57}$$

In figure 2, we show that for $\varepsilon = 2.5$ and 5, for example, agreement between $\partial\varphi/\partial x$ at $x = 0$ and the exact solution (57) is such that the curves for the solutions in each case overlap. $\partial\varphi/\partial x$ was evaluated using a simple forward difference, based on the nodal values of $\varphi(0) = 0$ and the interpolated value $\varphi(x_1)$, but could, of course, have been computed directly from using the Runge–Kutta method on (55) at $x = 0$. In order to assess the values of ε for which the solution to (53)–(55) is likely to be mass conserving, M_1 ($= \partial\varphi/\partial x$ at $x = 1$) was computed over some non-dimensional time interval $[0, T]$ using a simple backward difference formula, as explained above. For the results presented in figure 3, T was chosen equal to 10. This value was chosen because it was found that the computed $M_1(t)$ reached steady state at some $t < T$, at least for the values of Δx and ε considered here. Computations

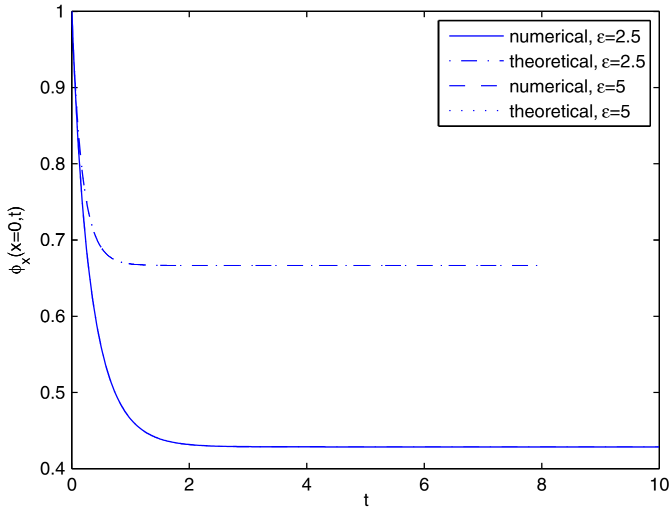

Figure 2. Comparison, for $\varepsilon = 2.5$ and 5, between $\partial\varphi/\partial x$ at $x = 0$ and the exact solution (57); $\Delta x = 0.0025$, $\Delta t = 0.00025$.

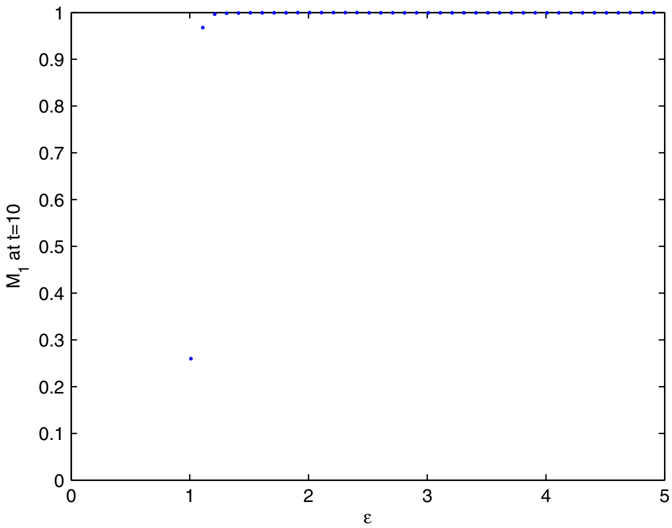

Figure 3. Extrapolated value of $M_1(T)$ against ε, $\Delta t = 0.0001$.

of M_1 for all the results that follow were performed with a dimensionless time step $\Delta t = 1 \times 10^{-4}$. Yet smaller values of Δt did not lead to any noticeable changes in the computed results.

In figure 3, we plot the values of $M_1(T)$ obtained by extrapolating to zero mesh size the linear least-squares polynomial based on computations of $M_1(T)$ with $\Delta x = 1 \times 10^{-3}, 2.5 \times 10^{-3}, 5 \times 10^{-3}$ and 1×10^{-2}. That $M_1(T)$ is visibly below 1 when $\varepsilon \lessapprox 1.43$ would appear to indicate that loss of mass conservation occurs before T over this range of ε. In figure 4, we show the approximate gel times t_{gel} as functions of ε and computed to be the smallest value of $t \leqslant T$ at which the extrapolated $M_1(t)$ dropped below 0.99 and 0.995. From the dimensionless form of (33) we know that when $\varepsilon = 0$, $t_{\text{gel}} = 0.5$, and this is borne out

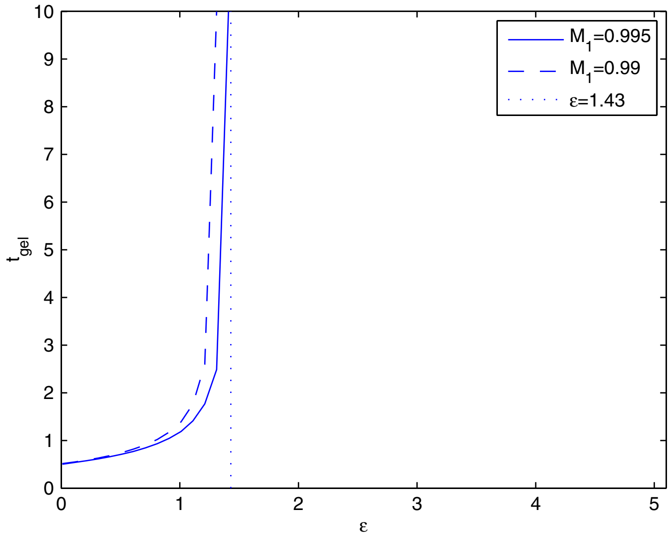

Figure 4. Approximate gel times t_{gel} against ε computed from the extrapolated value of M_1. $\Delta t = 0.0001$.

by the results shown in figure 4. Our experimental observations at higher values of ε await, however, confirmation from a rigorous mathematical analysis.

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