Nonscaling and source-induced scaling behaviour in aggregation model of movable monomers and immovable clusters

1991 J. Phys. A: Math. Gen. 24 4789

(http://iopscience.iop.org/0305-4470/24/20/014)

J. Phys. A: Math. Gen. 24 (1991) 4787-4803. Printed in the UK

Non-scaling and source-induced scaling behaviour in aggregation models of movable monomers and immovable clusters

N V Brilliantov† and P L Krapivsky‡

† Physics Department, Moscow State University, Moscow 119899, USSR
‡ Central Aerohydrodynamic Institute, Academy of Sciences of the USSR, 140160 Zhukovsky-3, Moscow region, USSR

Received 21 March 1991, in final form 24 June 1991

Abstract. General discussion of the aggregation kinetics for the wide class of aggregation models in which cluster growth occurs by bonding reactions between movable monomers and immovable clusters is presented. The study is carried out in terms of Smoluchowski's rate equations. We treat a general homogeneous case where the monomer–cluster reaction rates vary as k^γ, with the cluster size k. For models with $0<\gamma<1$ without a source we find that systems evolve to a final 'frozen' state. Evolution behaviour of the system appears to be non-scaling, but the deviation from a frozen state has a self-similar form. For the systems with a source, we have found that the solution has a scaling form in the most important part of the cluster-size distribution except for an asymptotically ignorable tail. We also carried out the analysis of the structure of the tail and of the thin boundary layer separating the scaling and non-scaling tail regions. The qualitative explanation of non-scaling and source-induced scaling behaviour may be made in terms of 'internal' time inherent for the models of these types.

1. Introduction

Aggregation is an irreversible physical process in which a number of basic units (monomers) stick together to build clusters (polymers). One of the main problems of aggregation kinetics is to describe the time evolution of cluster-size distribution (for a review see Drake 1972 or Ernst 1986). As a rule, the aggregation process is carried out in two stages. The first stage is a diffusive one: the clusters diffuse in a medium to meet one another, whereas at the second stage the bonding reaction occurs. In recent years, two limiting models for irreversible aggregation have been introduced. These are diffusion-limited cluster–cluster (CLCL) aggregation (Meakin 1983, Kolb *et al* 1983) and reaction-limited CLCL aggregation (Jullien and Kolb 1984, Kolb and Jullien 1984). The kinetics of such models was first discussed by Botet and Jullien (1984). A comprehensive review of this rapidly developing field is given by Meakin (1988). In a variety of aggregation processes the diffusion stage plays the crucial role. It was shown both analytically and numerically (see e.g. Kolb 1984, Meakin *et al* 1985) that the aggregation kinetics is strongly influenced by the cluster-size dependence of the diffusion coefficients.

The aim of our paper is to carry out a detailed analysis of the case when the diffusion coefficients of clusters are negligibly small (equal to zero in our model)

0305-4470/91/204787+17$03.50

compared with the diffusion coefficient of monomers. As we shall show, the kinetic behaviour in these systems drastically changes. Moreover, we have established that the evolution in systems with a source of monomers and source-free systems is principally different.

To describe the aggregation kinetics Smoluchowski equations are used (Smoluchowski 1917):

$$dC_k/dt = \tfrac{1}{2}\sum_{i+j=k} C_iC_jK_{ij} - C_k\sum_j C_jK_{kj}. \tag{1}$$

Here C_k is the concentration of clusters containing k monomer units (k-mers, $k=1,2,\ldots$), K_{ij} the rate constants for the reaction between i- and j-mers, producing $(i+j)$-mer. The gain and loss terms in the right-hand side of (1) describe the formation of k-mers of smaller clusters and the loss of k-mers due to their reaction with other monomers and clusters.

In the case of interest the cluster growth occurs only by means of addition of movable monomers to immovable clusters; cluster–cluster reactions are impossible in such systems. For this reason, the aggregation models of this type were called 'addition models' (Hendricks and Ernst 1984). For these models $K_{ij}=0$ when $i>1$ and $j>1$ and the rate kernel may be written as follows:

$$K_{ij} = S_i\delta_{j1} + S_j\delta_{i1}. \tag{2}$$

Equation (2) means that the K_{ij} matrix is 'hook-shaped' because its non-zero elements are only in the first column and first row. The kinetic equations (1) now attain the simple form

$$dC_1/dt = -S_1C_1^2 - C_1\sum_j S_jC_j + Q \tag{3}$$

$$dC_k/dt = C_1(S_{k-1}C_{k-1} - S_kC_k) \qquad k\geqslant 2.$$

Here the source term Q is added to account for the possible external source of monomers in a system. The kinetic equations of the 'addition' type (3) arise in a variety of problems. for example, an aggregation of point defects in solids is governed by (3) (Brilliantov and Krapivksy 1989). In this case the diffusion coefficients D_k of clusters, containing k point defects, are negligible compared with the diffusion coefficient for point defects D_1 (i.e. $D_k \ll D_1$ for $k>1$). Therefore, the rate matrix K_{ij} given by the Smoluchowski's formula

$$K_{ij} = 4\pi(R_i+R_j)(D_i+D_j) \tag{4}$$

(here R_j is the radius of j-mer), converts into the hook form (2) with

$$S_i = 4\pi(R_1+R_i)D_1. \tag{5}$$

Another example is an aggregation process caused by hydrolysis (Matsoukas and Gulary 1989). Hydrolysis produces the active monomers which subsequently react with either active monomers or polymeric species, containing i-monomers and producing $(i+1)$-mers. There are three irreversible steps in such a process:

$$C_0 \to C_1 \text{ (hydrolysis)} \qquad C_1 + C_1 \to C_2 \text{ (nucleation)}$$

$$C_i + C_1 \to C_{i+1} \text{ (growth)}.$$

The corresponding rate equations also have the 'addition' form:

$$\begin{aligned} \dot{C}_0 &= -S_0 C_0 \\ \dot{C}_1 &= S_0 C_0 - S_1 C_1^2 - C_1 \sum_j S_j C_i \\ \dot{C}_k &= C_1 (S_{k-1} C_{k-1} - S_k C_k) \qquad k \geqslant 2. \end{aligned} \tag{6}$$

In order to proceed with the solution of (3) and (6) we should choose a functional dependence of the growth reaction rate coefficients S_k on the cluster size k. It is natural to assume a power-law dependence of the form $S_k \propto k^\gamma$. This choice includes most physical situations. Moreover, we expect that the exponent γ lies in the interval $0 \leqslant \gamma \leqslant 1$. The reason is that the number of active sites on a cluster cannot increase faster than its size, i.e. that S_k/k is bounded as $k \to \infty$. The constant kernel ($\gamma = 0$) corresponds to a growth process with the rate independent of the particle size. Such is the case of the linear polymer growth (fixed number of reaction sites per particle). The proportional kernel ($\gamma = 1$) describes a process where all the monomers within a cluster (or a fixed fraction thereof) are the potential sites for growth. Real cases lie in between. As will be shown below, the parameter γ is the only one which defines the kinetic behaviour of the systems in a scaling region.

The exact value of parameter γ may be established on the basis of microscopic consideration of aggregation processes. However, some conclusions may be deduced for the most interesting cases of reaction- and diffusion-limited growth. In a reaction-limited process, the growth rate is limited by the bonding reactions. They are proportional to the number of possible bonding configurations between the monomer and cluster. So the growth kernel scales as the cluster surface area. Thus, for compact clusters in d-dimensional space, $\gamma = (d-1)/d$ whereas for fractal clusters $1 > \gamma > (d-1)/d$.

For diffusion-limited processes, Smoluchowski's formula (4) gives

$$S_k = 4\pi D_1 (R_1 + R_k) \sim R_k \sim k^{1/d_f} \qquad \text{at } k \gg 1. \tag{7}$$

This means that $\gamma = 1/d_f$ where d_f is the (fractal) dimension of clusters. For example, $\gamma = \frac{1}{3}$ for compact 3D clusters, $\gamma = \frac{1}{2}$ for compact disk-shaped clusters and $\gamma \approx 0.37$ for fractal clusters arising in a diffusion-limited growth (Witten and Sander 1981, Meaking 1988). Note that real diffusion-limited aggregation processes are much more complex and can involve additional factors such as reorganization after initial bonding step, long-range forces, rotational diffusion, etc. Some attempts have already been made to include some of these processes in simple aggregation models, see e.g. Meakin and Jullien (1985), Kolb (1986), Jullien (1985) and Meakin (1984, 1985). These additional factors may change fractal dimensionality of the aggregates. Thus, the γ exponent can be varied depending on the details of aggregation process.

It is interesting to note that the aggregation models of addition type are closely related to the diffusion-limited CLCL aggregation models with cluster diffusion coefficient varying as k^α at the limit $\alpha \to -\infty$ (k is the number of monomers in a cluster). The addition models describe the first stage (with shortest timescale) for the latter processes when only monomers are regarded as movable. When monomers are exhausted the second stage of the aggregation process starts. In the second stage dimers react with dimers and other clusters with $k > 2$. The timescale of the second stage is much greater than that of the first one, but aggregation kinetics is just the same with minor changes: $D_1 \to D_2$, $C_1 \to C_2$ and with initial conditions determined by the final

cluster-size distribution $C_k(t \to \infty)$ of the first stage of aggregation. When the second stage is completed the third stage starts, etc. So every stage of this hierarchical aggregation process may be regarded in terms of the addition model. The opposite case, $\alpha \to +\infty$, is also interesting (see e.g. Meakin *et al* 1985).

The outline of this paper is as follows. In section 2, we consider the addition models without the source. For the constant kernel and proportional kernel, the exact results have been found, while for the intermediate case $0 < \gamma < 1$ we have obtained asymptotic results in the limit $t \to \infty$ and $k \to \infty$.

We have shown that the solution of Smoluchowski's system with a hook-shaped rate matrix is non-scaling. This is in sharp contrast with the scaling behaviour of the most known solutions for the power-law kernels for an ordinary rate matrix with non-zero elements.

To complete the analysis, the unphysical case $\gamma > 1$ has also been investigated. It was established that gelation occurs in an infinitesimal time interval.

In section 3, the addition models with a source are considered. Surprisingly, the solutions we have found in the presence of a source fall into a scaling form. Thus we shall call this type of scaling a source-induced one.

To have a deeper insight into the kinetic behaviour of the system we introduce the concept of internal time. We show that the finiteness of the internal time is closely related to the absence of scaling in the system.

In section 4, we summarize our results and draw some conclusions.

2. Addition models without a source

The addition model with reaction rate matrix (2) and homogeneous reaction constants $S_k = Ak^\gamma$ hereafter will be referred to as the γ-hook model. Samsel and Perelson (1982) and Hendricks and Ernst (1984) have shown that 0-hook and 1-hook models are analytically solvable. We briefly sketch their results and concentrate on asymptotic behaviours (at $k \to \infty$ and $t \to \infty$) for these and for more general γ-hook models. In this section, we shall use the non-dimensional time t measured in units of $(AC_0)^{-1}$, where C_0 is the unit concentration.

2.1. 0-hook model

Defining the rescaled time variable

$$T = \int_0^t \mathrm{d}t'\, C_1(t') \tag{8}$$

we linearize the rate equations (3)

$$\frac{\mathrm{d}C_1}{\mathrm{d}T} = -C_1 - N \qquad \frac{\mathrm{d}N}{\mathrm{d}T} = -N \tag{9a}$$

$$\frac{\mathrm{d}C_k}{\mathrm{d}T} = C_{k-1} - C_k \qquad k \geqslant 2. \tag{9b}$$

Here $N = \Sigma_j C_j$ is the total number of clusters. It is easy to solve (9a) and then all (9b), recurrently. For the monodisperse initial data, $C_k(0) = \delta_{1k}$, we find

$$C_k(T) = \left[\frac{T^{k-1}}{(k-1)!} - \frac{T^k}{k!}\right] \exp(-T) \qquad k \geqslant 1. \tag{10}$$

Combining (8) and (10), one can easily find the relation between the 'physical' time t and the 'internal' time T:

$$t = \int_0^T \frac{dT'}{(1-T')} \exp(-T'). \tag{11}$$

Note that whereas t increases on an infinite time interval, $0 < t < \infty$, T increases on a finite time interval, $0 < T < T_f = 1$.

The asymptotic cluster-size distribution at $t \to \infty$ is the following:

$$C_k(\infty) = \frac{k-1}{ek!}. \tag{12}$$

In many recent studies, the scaling behaviour

$$C_k(t) = S^{-2}\Phi(k/S) \qquad S \sim t^z \tag{13}$$

had been observed in the scaling regime

$$k \to \infty \qquad t \to \infty \qquad k/S = \text{finite} \tag{14}$$

(for a review see Ernst 1986). The solutions of the 0-hook model do not belong to the scaling class.

One can give a qualitative explanation of this fact in terms of previously introduced internal time. This time is a physically inherent 'time' of the model and whereas it changes in a finite interval $0 < T < T_f$, the scaling kinetic behaviour has no 'time' to develop. The system therefore falls into the final state (12), which one can call the 'frozen' state. More careful analysis shows, however, that the deviation form (non-universal) frozen state has a self-similar form. Precisely, from (10) and (12) we find the asymptotic solution

$$\begin{aligned} &k \to \infty \qquad T \to T_f \qquad k(T_f - T) = x = \text{finite} \\ &C_k / C_k(\infty) \to \exp(-\lambda x) \qquad \lambda = 1. \end{aligned} \tag{15}$$

Similar results may be found for the 0-hook hydrolysis model. Omitting the details of straightforward calculations we give the final results:

$$C_1 \to (N_0 - N_1 T)\exp(-T) \qquad t \to \infty \tag{16}$$

$$C_{k+1} \to \frac{C_1(0)}{k!}\exp(-T)T^k \qquad t \to \infty \qquad k \gg 1 \tag{17}$$

$$N \to N_1 \exp(-T) \qquad t \to \infty. \tag{18}$$

Here N_0 and N_1 are some non-universal constants depending on initial conditions, T is the internal time:

$$t = \int_0^T \frac{dT'}{(N_0 - N_1 T')} \exp(-T'). \tag{19}$$

Again we see that the internal time changes in a finite interval $0 < T < T_f = N_0/N_1$ and that the deviation from freezing state has the self-similar form (15) with $\lambda = T_f^{-1}$.

2.2. 1-hook model

Now we turn to the 1-hook model:

$$\frac{dC_1}{dT} = -C_1 - M \qquad M = 1 \tag{20a}$$

$$\frac{dC_k}{dT} = (k-1)C_{k-1} - kC_k \qquad k \geqslant 2. \tag{20b}$$

Here $M = \Sigma_j jC_j$ is the total mass. Introducing the generating function

$$g = \sum_j C_j(T)\exp(jz) \tag{21}$$

we deduce from (18) and (19)

$$\frac{\partial g}{\partial T} = (e^z - 1)\frac{\partial g}{\partial z} - e^z \tag{22}$$

or

$$\left(\frac{\partial}{\partial T} - \frac{\partial}{\partial \xi}\right)[g + \log(1 - e^{-\xi})] = 0 \tag{23}$$

where

$$\xi = \int dZ(e^z - 1)^{-1} = \log(1 - e^{-z}). \tag{24}$$

One can easily find an exact solution of (23) with arbitrary initial data. For the monodisperse ones, we obtain

$$g(\xi, t) = [1 - \exp(\xi + T)]^{-1} + \log\left[\frac{1 - \exp(-\xi - T)}{1 - \exp(-\xi)}\right] \tag{25}$$

or

$$C_k(T) = e^{-T}(1 - e^{-T})^{k-1} - k^{-1}(1 - e^{-T})^k. \tag{26}$$

In terms of 'physical' time we have

$$C_k(t) = [(1 - e^{-t})^{k-1} - k^{-1}(1 - e^{-t})^k](2 - e^{-t})^{-k}. \tag{27}$$

As t increases from zero to infinity, the internal time $T = \log(2 - e^{-t})$ increases on the finite time interval, $0 < T < T_f$, with $T_f = \log 2$. The deviation from non-universal frozen state ($C_k(\infty) = (1 - k^{-1})2^{-k}$ for the monodisperse initial data) again may be recast into the scaling form (15) with $\lambda = 1$.

Similar results can be found for the 1-hook model with hydrolysis. We have a simple relation for $C_0(t)$:

$$\dot{C}_0 = -aC_0 \qquad C_0 = \exp(-at). \tag{28}$$

Here $a = S_0$ is the reaction rate of hydrolysis. The monomer concentration may be found from the second equation of the system (6)

$$\dot{C}_1 + C_1(C_1 + M_0 + 1 - e^{-at}) = a\,e^{-at} \tag{29}$$

where M_0 is the initial mass, $M_0 = \Sigma_j jC_j(0)$, and the concentrations of k-mers may be found by means of simple recursion

$$C_{k+1}(T) = k \int_0^T \mathrm{d}u\, C_1(T-u) \exp(-2u)[1-\exp(-u)]^{k-1}. \tag{30}$$

Equations (29) and (30) may be treated analytically for particular initial conditions: $C_k(0) = 0$ at $k > 1$, $C_1(0) = 2a - 1$. One obtains in this case for the concentration of monomers:

$$C_1 = \exp(-at) + A(t)\left[1 + \int_0^t A(\tau)\,\mathrm{d}\tau\right]^{-1} \tag{31a}$$

where the function $A(t)$ is defined as

$$A(t) = 2(a-1)\exp[-2at - (1-\mathrm{e}^{-at})/a]. \tag{31b}$$

The simplest solution corresponds to $a = 1$:

$$C_1(t) = \exp(-t) = 1 - T. \tag{32}$$

We see that the internal time again changes in a finite time interval, $0 < T < T_f = 1$. For the asymptotic behaviour we obtain

$$C_{k+1} \simeq C_1(0)\,\mathrm{e}^{-2T}(1-\mathrm{e}^{-T})^k \qquad k \gg 1 \tag{33}$$

and once more deduce a self-similar form (15) with $\lambda = [\exp(T_f) - 1]^{-1}$ for the deviation from the frozen state distribution.

2.3. *γ-hook model with $0 < \gamma < 1$*

In the case of intermediate γ, $0 < \gamma < 1$, we investigate the system of equations

$$\frac{\mathrm{d}C_1}{\mathrm{d}T} = -C_1 - M_\gamma \qquad M_\gamma = \sum_{k=1}^{\infty} k^\gamma C_k \tag{34a}$$

$$\frac{\mathrm{d}C_k}{\mathrm{d}T} = (k-1)^\gamma C_{k-1} - k^\gamma C_k \qquad k \geqslant 2. \tag{34b}$$

For the monodisperse initial data we find a lower and upper bound for the functions C_k and M_γ. The equation for the momentum function M_γ may be derived from (34):

$$\frac{\mathrm{d}M_\gamma}{\mathrm{d}T} = -M_\gamma + \sum_{k=1}^{\infty} C_k k^\gamma[(k+1)^\gamma - k^\gamma]. \tag{35}$$

First, we find a lower bound for $M_\gamma(T)$:

$$\frac{\mathrm{d}M_\gamma}{\mathrm{d}T} \geqslant -M_\gamma \qquad M_\gamma \geqslant \exp(-T) \tag{36}$$

and then an upper bound for $C_1(T)$:

$$\frac{\mathrm{d}C_1}{\mathrm{d}T} + C_1 \leqslant -\mathrm{e}^{-T} \qquad C_1 \leqslant (1-T)\,\mathrm{e}^{-T}. \tag{37}$$

From (37) it follows that the internal time changes in a finite time interval $0 < T < T_f$ and we have an upper bound for T_f, $T_f \leqslant 1$.

Using the obvious inequality

$$(k+1)^\gamma \leqslant k^\gamma + \gamma k^{\gamma-1} \leqslant k^\gamma + \gamma \tag{38}$$

we derive an upper bound for $M_\gamma(T)$

$$\frac{dM_\gamma}{dT} \leqslant -(1-\gamma)M_\gamma \qquad M_\gamma \leqslant \exp[-(1-\gamma)T] \tag{39}$$

and then a lower bound for $C_1(T)$

$$\frac{dC_1}{dT} + C_1 \geqslant -e^{-(1-\gamma)T} \qquad C_1 \geqslant \left[1+\frac{1}{\gamma}\right] e^{-T} - \frac{1}{\gamma} e^{-(1-\gamma)T}. \tag{40}$$

Further, from (40) we deduce a lower bound for T_f, $T_f \geqslant (1/\gamma)\log(1+\gamma)$, which coincides with the exact results $T_f = 1$ and $T_f = \log 2$ in the limiting cases $\gamma = 0$ and $\gamma = 1$, respectively.

Using (34*b*) one finds the relation

$$C_{k+1}(T) = k^\gamma \int_0^T du\, C_1(T-u) \exp[-(k+1)^\gamma u]. \tag{41}$$

We suppose that the deviation from frozen state has a self-similar form (15) again. Under this assumption one can estimate a frozen state for large k:

$$C_{k+1}(\infty) \cong k^\gamma C_k(\infty) \int du \exp[-ku - (k+1)^\gamma u]$$

$$\approx C_k(\infty) k^{\gamma-1}(1-k^{\gamma-1}) \qquad \text{at } k \gg 1. \tag{42}$$

Using this result together with a large-k approximation, $\Pi_{n=1}^{k}[1-n^{\gamma-1}] \propto \exp(-k^\gamma/\gamma)$, we obtain

$$C_{k+1} \propto \frac{\exp(-k^\gamma/\gamma)}{(k!)^{1-\gamma}}. \tag{43}$$

Up to an overall numerical factor this asymptotic solution for the γ-hook model, $0 < \gamma < 1$, is independent of the initial conditions. Similar results may be found for the γ-hook model with hydrolysis.

2.4. Gelation in the hook models with $\gamma > 1$

It is well known that in some aggregation processes gelation take place. Mathematically, the sol-gel transition is manifested by violation of the mass-conservation law after some time interval t_g (gelation time). Physically, it means that a huge cluster, spreading throughout the system is created at $t \geqslant t_g$. The gelation kinetics critically depends on the functional form of the rate kernel. From the preceding analysis it follows that gelation for the hook kernels may occur only when $\gamma > 1$. Therefore the question arises: does gelation occur on our model and what is the value of the time t_g? The second question is a non-trivial one accounting the recent speculations of various authors who stated that for certain forms of the reaction kernels gelation may take place instantaneously. Analytical arguments supporting this view have been given by Hendricks *et al* (1982) and by Ziff (1984). Different evidence, supporting the possible occurrence of an instantaneous gelation transition, comes from Monte Carlo simulations (Domilovskii *et al* 1978, Spouge 1985). Recently van Dongen (1987) confirmed the

occurrence of a gelation transition within infinitesimal time for some homogeneous reaction kernels.

In our hook model with $\gamma > 1$ instantaneous gelation indeed takes place (i.e. $t_g = 0$). We have found the structure of post-gel solution: $C_1(T) = 0$; $C_k(T) = C_k(0)$, for all $k > 1$ and $t > 0$. These results were found following the lines of van Dongen (1987).

3. Addition models with a source

A source-enhanced cluster-cluster aggregation can be observed in various natural phenomena such as atmospheric aerosols (Klett 1975, Friedlander 1977, White 1982, Crump and Seinfeld 1982), star formation (Field and Saslaw 1965), the formation of inter-stellar dust grains (Salpeter 1977, Hayakawa and Hayakawa 1988), vapour-deposited thin films (Family and Meakin 1989), the diffusion-limited growth of surface structures (Brilliantov *et al* 1989), diffusion-controlled deposition (Racz and Vicsek 1983, Matsushita and Meaking 1988) and radiation-induced segregation (Bourgoin and Corbett 1978, Corbett 1979). These systems have been investigated theoretically in the mean-field approximation, i.e. using Smoluchowski's coagulation equation, by Kayakawa (1987); numerically by Vicsek *et al* (1985), Racz (1985a), Meakin *et al* (1985) and Hayakawa *et al* (1987); and exactly in 1D (Racz 1985b, Takayasu *et al* 1988, Takayasu 1989, Takayasu and Takayasu 1989, Thompson 1989, Doering and ben-Avraham 1989, Cheng *et al* 1989, Meakin 1989).

In many of these systems the increase in the total mass of cluster concentration is balanced by the continual coalescence of clusters. The concentration of clusters of any fixed mass reaches a steady state and the cluster-mass distribution follows power law with infinite variance.

In this section we discuss the hook models with a source. As we shall show, there are no stationary states in these models. Furthermore, the kinetic behaviour at long time exhibits rather peculiar scaling. This is in sharp contrast with the kinetic behaviour of usual aggregation models with a source.

3.1. 0-hook model

For a 0-hook model with a source, we obtain the following rate equations:

$$\frac{dC_1}{dt} = 1 - C_1(C_1 + N) \qquad \frac{dN}{dt} = 1 - C_1 N \tag{44a}$$

$$\frac{dC_k}{dT} = C_{k-1} - C_k \qquad k \geq 2. \tag{44b}$$

Here the non-dimensional time t is measured in units of $(AQ)^{-1/2}$ and the non-dimensional concentrations in units of $(Q/A)^{1/2}$, where Q represents the rate of monomer production by an external source.

Equations (44*a*) yield the following closed equation for $C_1(t)$:

$$\ddot{C}_1 + \dot{C}_1^2/C_1 + \dot{C}_1/C_1 + 2\dot{C}_1 C_1 + C_1^3 = 0 \tag{45}$$

which can be used to obtain the asymptotic decay

$$C_1 \to (3t)^{-1/3} \qquad \text{at } t \to \infty. \tag{46}$$

Combining the definition of the internal time (8) and the asymptotic solution (46) one finds the relation between the physical and internal times:

$$T \to (1/2)(3t)^{2/3} \qquad \text{at } t \to \infty \tag{47}$$

i.e. $C_1(T) \to (2T)^{-1/2}$ at $t \to \infty$. Note that the internal time now varies in an infinite time interval, $0 < T < \infty$. This contrasts with the behaviour in the hook models without a source and indicates that scaling behaviour may be expected.

To explain the behaviour of the solution at $k \gg 1$, we replace the difference operator on the right-hand side of (44*b*) by a differential operator and solve the resultant wave equation

$$\left(\frac{\partial}{\partial T}+\frac{\partial}{\partial k}\right)C_k(T) \approx 0 \qquad C_k(T) \approx C_1(T-k) = [2T(1-k/T)]^{-1/2}. \tag{48}$$

This result may be derived more rigorously. Using a recursive method to solve (44*b*), we obtain

$$C_{k+2}(T) = \frac{1}{k!}\int_0^T \mathrm{d}u\, C_1(T-u)u^k \exp(-u). \tag{49}$$

First, we consider a behaviour of the exact solution (49) in the scaling region

$$T \to \infty \qquad k \to \infty \qquad x = k/T = \text{fix} < 1. \tag{50}$$

One can estimate the integral (49) by the Laplace method which yields

$$C_{k+2}(T) \approx \frac{1}{k!}\exp(k \log k - k)\int_{-\infty}^{\infty} \mathrm{d}u\, C_1(T-k+u)\exp(-u^2/2k) \approx C_1(T-k). \tag{51}$$

This result agrees with the earlier result (48).

Second, we consider a behaviour of the solution (49) in the region

$$k \to \infty \qquad T \to \infty \qquad \text{and} \qquad (k-T)/T \ll 1. \tag{52}$$

A simple analysis shows that very rapid decrease of the function $C_k(T)$ occurs in a 'layer' of order $\sim T^{1/2}$. Therefore it is natural to introduce the 'layer' variable:

$$y = \frac{k-T}{T^{1/2}}. \tag{53}$$

Then we transform (49) into the following self-similar expression:

$$C_k(T) \approx (2T)^{1/4} f(y)$$

$$f(y) = (2\pi)^{-1/2}\int_0^{\infty} \mathrm{d}u \exp[-(u+y)^2]u^{-1/2}. \tag{54}$$

In the last region $x > 1$, a main contribution to the integral (49) is accumulated near the upper limit:

$$C_k(T) = \frac{1}{k!}T^k \exp(-T)F(x)$$

$$F(x) = \int_0^T \mathrm{d}u\, C_1(T-u)\exp[-u(x-1)] \qquad x = k/T. \tag{55}$$

Summing all these results, we finally arrive at the expression

$$C_k(T)=\begin{cases}(2T)^{-1/2}(1-x)^{-1/2} & \text{at } x<1 \qquad x=k/T \\ (2T)^{-1/4}f(y) & \text{at } x\approx 1 \qquad k=T-(2T)^{1/2}y \\ T^{k-2}\exp(-T)[(k-2)!]^{-1}F(x) & \text{at } x>1.\end{cases} \tag{56}$$

Thus we see that the solution has the scaling form at $x<1$, the self-similar form at $x=1$ and $-\infty<y<\infty$ and the non-scaling form at $x>1$, with non-universal factor $F(x)$ depending on initial conditions. So we conclude that the behaviour of this model is rather peculiar. Similar results have also been found for the 0-hook model with hydrolysis.

3.2. 1-hook model

Now we turn to 1-hook model with a source, which is described by the following rate equations:

$$\frac{dC_1}{dt}=1-C_1(C_1+M) \tag{57a}$$

$$\frac{dC_k}{dT}=(k-1)C_{k-1}-kC_k \qquad k\geqslant 2. \tag{57b}$$

The mass conservation gives $M=M_0+t$, i.e. (57*a*) is the well known Riccati equation. It is easy to check that $C_1=(M_0+t)^{-1}$ is the particular solution of (57*a*). Consequently, one can construct an exact solution with an arbitrary initial condition on $C_1(0)$.

To proceed further we use the method of generating functions and obtain the equation

$$\partial g/\partial T=(\exp(z)-1)\partial g/\partial z+C_1(T)\exp(2z) \tag{58}$$

for the generating function

$$g(T,z)=\sum_{k=2}^{\infty}C_k(T)\exp(kz). \tag{59}$$

Solving (58) yields

$$g=\int_0^T dT_1\, C_1(T_1)[1-\exp(T-T_1+\xi)]^{-2} \tag{60}$$

where ξ is given by (24). Expanding (60) we obtain the exact solution

$$C_k(T)=(k-1)\int_0^T du\, C_1(T-u)\exp(-2u)[1-\exp(-u)]^{k-2}. \tag{61}$$

In the scaling region

$$k\to\infty \qquad T\to\infty \qquad x=k\exp(-T)=\text{fix} \tag{62}$$

we come to the scaling-like form of (61):

$$C_k(T)=\exp(-T)\Phi(x) \tag{63}$$

$$\Phi(x)=x\int_0^{\infty}du\, C_1(u)\exp(2u-x\,e^u).$$

Here C_1 is the solution of the Riccati equation (57a) written in terms of the internal time T. When $x \ll 1$ (63) yields a true scaling behaviour

$$C_k(t) \simeq 1/t \qquad \text{at } k \ll t. \tag{64}$$

In contrast, at moderate $x \geqslant 1$ (i.e. at $k \geqslant t$) $\Phi(x)$ crucially depends on the initial condition $C_1(0)$. Such a non-universality becomes especially visible at $x \gg 1$ where one can find

$$\Phi(x) \simeq C_1(0) \exp(-x) \qquad \text{when } C_1(0) > 0 \tag{65a}$$

and

$$\Phi(x) \simeq (\pi/2x)^{1/2} \exp(-x) \qquad \text{when } C_1(0) = 0. \tag{65b}$$

In closing this subsection we note that for the simplest case $C_1(0) = 1$ we succeed in finding the exact solution

$$C_k(t) = t^{k-1}/(1+t)^k. \tag{66}$$

Now we turn to a general case of a γ-hook model with a source. We have observed three different types of kinetic behaviour in the regions $0 < \gamma < \frac{1}{2}$, $\frac{1}{2} < \gamma < 1$ and $\gamma = 1$. In the following subsections we present these results.

3.3. γ-hook models with $0 < \gamma < \frac{1}{2}$

The rate equations for the γ-hook model with a source are the following:

$$\frac{dC_1}{dT} = 1 - C_1(C_1 + M_\gamma) \qquad M_\gamma = \sum_{k+1}^{\infty} k^\gamma C_k \tag{67a}$$

$$\frac{dC_k}{dT} = (k-1)^\gamma C_{k-1} - k^\gamma C_k \qquad k \geqslant 2. \tag{67b}$$

As was demonstrated above, the asymptotic decay of the functions $C_k(t)$ have the power-law form $C_k(t) \propto t^{-\beta}$ at $t \to \infty$, where $\beta = \frac{1}{3}$ for $\gamma = 0$ and $\beta = 1$ for $\gamma = 1$ (see (46) and (64)). Therefore it is natural to assume that the solution for an arbitrary γ, $0 < \gamma < 1$, has the same form with a parameter β in the interval $\frac{1}{3} < \beta < 1$. For $\beta < 1$ we find that $dC_k/dt \ll C_1 C_k$ as $t \to \infty$ and the quantities dC_k/dt can be neglected in the first approximation. As a result we obtain from (67) the following asymptotic relations for sufficiently small k:

$$C_k(t) \to B k^{-\gamma} t^{-\beta} \qquad \Gamma(t) \to t^\beta / B \tag{68}$$

where $\Gamma(t) = M_\gamma(t)$.

To determine the constants B and β, we shall use the internal time T again. In terms of T we rewrite (68) as follows:

$$\begin{aligned} &T \to B(1-\beta)^{-1} t^{1-\beta} \\ &C_k(T) \to B^{1/(1-\beta)} (1-\beta)^{-\beta/(1-\beta)} T^{-\beta/(1-\beta)} k^{-\gamma} \\ &\Gamma(t) \to \{C_1(T)\}^{-1}. \end{aligned} \tag{69}$$

To find the cluster-mass distribution we write the kth rate equation (67) in the 'wave' form

$$(\partial/\partial T + \partial/\partial K)(k^\gamma C_k) = 0 \tag{70}$$

where we have rescaled the index variable

$$K = k^{1-\gamma}/(1-\gamma). \tag{71}$$

Solving (70) yields

$$C_k(T) = k^{-\gamma} f(T-K). \tag{72}$$

Combining (69) and (72) we find the function f and finally obtain

$$C_k(T) = B^{1/(1-\beta)}(1-\beta)^{-\beta/(1-\beta)}(T-K)^{-\beta/(1-\beta)}k^{-\gamma}. \tag{73}$$

This scaling result is valid in the scaling region

$$k \to \infty \qquad t \to \infty \qquad x = K/T = \text{fix} < 1. \tag{74}$$

Substituting (73) into the expression $\Gamma(T) = M_\gamma(T)$ and replacing the sum by the integral one finds

$$\Gamma(T) \simeq \int_1^\infty \mathrm{d}k\, k^\gamma C_k(T)$$

$$\simeq [B/(1-\beta)^\beta]^{1/(1-\beta)}(1-\gamma)^{\gamma/(1-\gamma)} \int_0^T \mathrm{d}K\, K^{\gamma/(1-\gamma)}(T-K)^{-\beta/(1-\beta)}. \tag{75}$$

The integral (75) converges for $\beta < \frac{1}{2}$. In this region (75) becomes

$$\Gamma \simeq [B/(1-\beta)^\beta]^{1/(1-\beta)}(1-\gamma)^{\gamma/(1-\gamma)}\Gamma\left(\frac{1}{1-\gamma}\right)\Gamma\left(\frac{1-2\beta}{1-\beta}\right)$$

$$\times\left[\Gamma\left(\frac{1}{1-\gamma}+\frac{1-2\beta}{1-\beta}\right)\right]^{-1} T^{1+\gamma/(1-\gamma)-\beta/(1-\beta)}. \tag{76}$$

Comparing this result with the expression for $\Gamma(T)$ in (69), we obtain

$$\beta = (3-2\gamma)^{-1} \tag{77a}$$

$$B = \frac{(1-\beta)^\beta}{(1-\gamma)^{\beta\gamma}}\left[\Gamma\left(\frac{3/2-\gamma}{1-\gamma}\right)\left(\Gamma\left(\frac{1}{1-\gamma}\right)\Gamma\left(\frac{1-2\gamma}{2-2\gamma}\right)\right)^{-1}\right]^{-\beta(1-\gamma)} \tag{77b}$$

which indicates that a self-consistency condition, $\beta < \frac{1}{2}$, imposes a restriction on the parameter γ, viz $0 \leqslant \gamma < \frac{1}{2}$. We also present our results for the moment function in this interval of γ:

$$M_\alpha(t) = \left(\frac{B}{2\beta}\right)^{(\alpha-1)/(1-\gamma)}\Gamma\left(\frac{5-4\gamma}{2-2\gamma}\right)\Gamma\left(\frac{\alpha-\gamma+1}{1-\gamma}\right)$$

$$\times\left[\Gamma\left(\frac{2-\gamma}{1-\gamma}\right)\Gamma\left(\frac{2\alpha+3-4\gamma}{2-2\gamma}\right)\right]^{-1} t^{(2\alpha-2\gamma+1)/(3-2\gamma)}. \tag{78}$$

Rigorously speaking, the solution (73) is valid only in the region ($k \to \infty$, $T \to \infty$, $x < 1$). Nevertheless, the results are valid asymptotically because the functions $C_k(T)$ decrease very rapidly when $x > 1$ and do not contribute to the integral (75) significantly. To describe the details of the behaviour of $C_k(T)$ in the vicinity of $k = T$ where $C_k(T)$ diverges in the continuum approximation (70), we would keep the high-order terms in the expansion of the difference operator in the right-hand side of (67b). We have found that the 'wavefront' of the cluster-size distribution located at $x = 1$ has a self-similar form:

$$C_k(T) = T^{-(1-2\gamma)/((2-2\gamma)^2)} f(y) \qquad y = (K-T)T^{-(1-2\gamma)/(2-2\gamma)} \qquad x \approx 1 \tag{79}$$

with some non-singular function $f(y)$. Note that the dependence $C_k(T)$ has a sharp peak as for the 0-hook model. The qualitative dependence of cluster concentration C_k versus cluster size k for $T \gg 1$ and $k \gg 1$ is presented in figure 1.

3.4. *Hook models with* $\gamma \geqslant \frac{1}{2}$

In the case $\gamma \geqslant \frac{1}{2}$ the integral in (75) includes a non-physical divergence at the upper limit. This is related to the use of the divergent expression (73) for $C_k(T)$ in the region $T-K \to 0$. As we have shown for the case $0<\gamma<\frac{1}{2}$, in this 'wavefront' region the solution converts into a non-singular form. Since we do not know the precise behaviour of $C_k(T)$ in this region, we can avoid this difficulty by introducing a cut-off in the upper limit of the integral (75). For $\gamma \leqslant \frac{1}{2}$ this does not modify the final result, whereas an unknown constant appears for $\gamma > \frac{1}{2}$ without modifications of the time dependence. Estimating the integral with the cut-off at the upper limit and comparing the result with (69) for $\Gamma(T)$, we find that in the interval $\frac{1}{2}<\gamma<1$ the following relation for the exponent β holds: $\beta = \gamma$. Therefore at $t \to \infty$ we obtain

$$C_k(T) \propto k^{-\gamma} T^{-\gamma/(1-\gamma)} \qquad T \propto t^{1-\gamma} \tag{80}$$

and for $x \leqslant 1$ $(x = k/T)$ one can use the expressions (71) and (72) again.

The reasoning becomes rather more complicated for $\gamma = \frac{1}{2}$. Assume that $C_1(T) = f(T)$. Then one finds

$$\Gamma(T) = \{C_1(T)\}^{-1} \tag{81a}$$

$$C_k(T) = k^{-1/2} f(T-K) \tag{81b}$$

where $K = 2k^{1/2}$. We shall seek the function $f(x)$ in the form $f(x) = [b(x)x]^{-1}$ treating $b(x)$ as a slowly varying function. This choice of the function f is prescribed by the

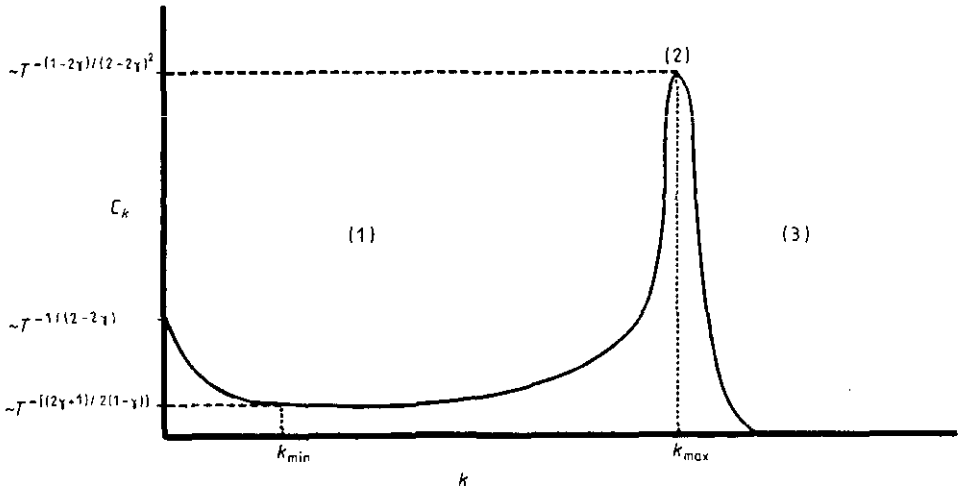

Figure 1. Qualitative dependence of cluster concentrations C_k versus cluster size k for $T \gg 1$ and $k \gg 1$ (T = internal time, $0 \leqslant \gamma \leqslant \frac{1}{2}$). Here $k_{max} = [(1-\gamma)T]^{1/(1-\gamma)}$, $k_{min} = k_{max}[2\gamma/(2\gamma+1)]^{1/(1-\gamma)}$. There are three different regions on the cluster-size distribution: (1) scaling region which contains the main part of the cluster distribution; (2) sharp self-similar peak centred at k_{max}, with the width $\simeq T^{(1-2\gamma)/(2-2\gamma)}$; (3) non-scaling front.

form of the solution (73). In the new notation we can write $\Gamma(T)$ as follows:

$$\Gamma(T) \simeq \frac{1}{2}\int_0^\infty \mathrm{d}x \frac{(T-x)}{xb(x)} = \frac{T}{2}\int_0^T \frac{\mathrm{d}x}{xb(x)} - \frac{1}{2}\int_0^T \frac{\mathrm{d}x}{b(x)}. \tag{82}$$

We assume that the second term on the right-hand side of (82) can be neglected at $T \gg 1$ compared with the first term. Using (81*a*) we therefore have

$$b(T) = \frac{1}{2}\int_0^T \frac{\mathrm{d}x}{xb(x)}. \tag{83}$$

Differentiating this equation and then solving the resultant equation we obtain

$$b(T) = (\log T)^{1/2}. \tag{84}$$

The validity of our assumption about the respective values of the terms in the left-hand side of (82) can now be verified by direct calculations. When $b(x)$ is known, one can determine all the relevant quantities:

$$\begin{aligned} &C_k(T) \simeq k^{-1/2}(\log T)^{-1/2}T^{-1} && \text{at } k \ll T \\ &T \simeq 2t^{1/2}(2\log t)^{1/4} && \text{at } t \to \infty. \end{aligned} \tag{85}$$

In terms of the physical time t we have

$$C_k(t) \simeq k^{-1/2}(\log t)^{-1/4}t^{-1/2} \qquad \text{at } k \ll t \tag{86a}$$

$$M_\alpha(t) \simeq t^\alpha (2\log t)^{(1-\alpha)/2} \qquad \text{at } t \to \infty. \tag{86b}$$

It should be noted that the relations (85) and (86) hold up to terms of the order of $\log(\log t)/\log t$, i.e. the solutions tend to the asymptotic expressions extremely slowly at $\gamma = \frac{1}{2}$.

In closing the section we sum the results for γ-hook models. In the scaling region $T \to \infty$, $k \to \infty$ and $x = k/T = \text{fix}$, the scaling behaviour is observed at $x < 1$, i.e.

$$C_k(t) \propto k^{-\gamma}t^{-\beta}\Phi(x) \qquad x = \frac{k}{t^z}. \tag{87}$$

Here the scaling exponents are

$$\beta = \begin{cases} (3-2\gamma)^{-1} & \text{at } 0 \leqslant \gamma \leqslant \frac{1}{2} \\ \gamma & \text{at } \frac{1}{2} < \gamma < 1 \end{cases} \tag{88}$$

and

$$z = \begin{cases} 2(3-2\gamma)^{-1} & \text{at } 0 \leqslant \gamma \leqslant \frac{1}{2} \\ 1 & \text{at } \frac{1}{2} < \gamma < 1. \end{cases} \tag{89}$$

The scaling function is given by the expression

$$\Phi(x) = \begin{cases} [1 - \text{constant } x^{1-\gamma}]^{-1/(2-2\gamma)} & \text{at } 0 \leqslant \gamma \leqslant \frac{1}{2} \\ [1 - \text{constant } x^{1-\gamma}]^{-\gamma/(1-\gamma)} & \text{at } \frac{1}{2} < \gamma < 1. \end{cases} \tag{90}$$

Note that the values $\gamma = \frac{1}{2}$ and $\gamma = 1$ play the role of critical points in the hook models which separate the different types of kinetic behaviours. At these points novel and rather unexpected behaviours are observed, viz log-correction factors for $\gamma = \frac{1}{2}$ and stretched wavefront for 'pre-gel' model $\gamma = 1$.

4. Summary

We have studied aggregation kinetics of addition models. These models describe the aggregation processes when the clusters are immovable whereas the monomers move diffusively. Kinetics of these models was investigated on the basis of the rate equations. This is an approximation of a mean-field type because it ignores the spatial fluctuations of the species concentrations.

The rate kernel matrix for addition models have a hook-shaped form. We have considered the power-law dependence of the rate constants. We observed that the kinetic behaviour crucially depends on the presence of a source in the system and on the power exponent γ of the rate constants.

For unphysical models with $\gamma > 1$, instantaneous gelation occurs. For models with $0 \leqslant \gamma \leqslant 1$, we found that in the systems without a source an asymptotic frozen state is achieved as time tends to infinity. The approach to a frozen state appears to be non-scaling. However, the deviation from a frozen state has a self-similar form. This contrasts with kinetic behaviours of most of the previously studied aggregation models with homogeneous kernels where scaling behaviour was observed.

For the systems with a source, we found that the solution has a scaling form in the most important part of the cluster-size distribution except for an asymptotically ignorable tail. We also carried out analysis of the tail structure and the thin boundary layer separating the scaling and non-scaling tail regions. Overall cluster-size distribution proves to be peak-shaped. We have estimated the location and width of the peak.

Our analysis has shown that two values of the power exponent γ, $\gamma = \frac{1}{2}$ and $\gamma = 1$, are somewhat critical points in the hook models. These points separate the regions with different kinetic behaviours. For the pre-gel point $\gamma = 1$, the stretched wavefront of cluster-size distribution has been observed, and for $\gamma = \frac{1}{2}$, we have found rather subtle behaviour with the logarithmic corrections to the usual power-law behaviour.

The established properties of the whole class of hook models may be qualitatively explained in terms of the internal time which is physically inherent for such models. For the systems without a source, it varies in a finite time interval. So the scaling has not time to develop. In contrast, in the systems with a source the internal time varies in an infinite time interval, and the scaling, although rather peculiar, does develop.

References

Botet R and Jullien R 1984 *J. Phys. A: Math. Gen.* **17** 2517
Bourgoin J C and Corbett J W 1978 *Rad. Eff.* **36** 157
Brilliantov N V and Krapivsky P L 1989 *Sov. Phys. Solid. State* **31** 271
Brilliantov N V, Volskii and Krapivksy P L 1989 *Sov. Phys. Surface* **12** 30
Cheng Z, Redner S and Leyvraz F 1989 *Phys. Rev. Lett.* **62** 2321
Corbett J W 1979 *Surf. Sci.* **90** 205
Crump J G and Seinfeld J H 1982 *J. Coll. Interface Sci.* **90** 469
Doering C R and ben-Avraham D 1989 *Phys. Rev. Lett.* **62** 2563
Domilovskii E R, Lushnikov A A and Piskunov V V 1978 *Dokl. Chem. Phys.* **240** 108
Drake R L 1972 *Topics of Current Aerosol Research* vol III part 2 ed G M Hidy and J R Brook (Oxford: Pergamon)
Ernst M H 1986 *Fractals in Physics* ed L Pietronero and E Tosatti (Amsterdam: Elsevier) p 289
Family F and Meakin P 1989 *Phys. Rev.* A **40** 3836
Field G B and Saslaw W C 1965 *Astrophys. J.* **142** 568
Friedlander S K 1977 *Smoke, Dust and Haze: Fundamentals of Aerosol Behavior* (New York: Wiley)

Hayakawa H 1987 *J. Phys. A: Math. Gen.* **20** L801
Hayakawa H and Hayakawa S 1988 *Publ. Astron. Soc. Japan* **40** 341
Hayakawa H, Yamamoto M and Takayasu H 1987 *Progr. Theory. Phys.* **78** 1
Hendricks E M and Ernst M H 1984 *J. Coll. Interface Sci.* **97** 176
Hendricks E M, Ernst M H and Ziff R M 1982 *J. Stat. Phys.* **31** 519
Jullien R 1985 *Phys. Rev. Lett.* **55** 1697
Jullien R and Kob M 1984 *J. Phys. A: Math. Gen.* **17** L639
Klett J D 1975 *J. Atmos. Sci.* **32** 380
Kolb M 1984 *Phys. Rev. Lett.* **53** 1653
—— 1986 *J. Phys. B: At. Mol. Phys.* **19** L263
Kolb M, Botet R and Jullien R 1983 *Phys. Rev. Lett.* **51** 1123
Kolb M and Jullien R 1984 *J. Physique Lett.* **45** L977
Matsoukas T and Gulary E 1989 *J. Coll. Interface Sci.* **132** 13
Matsushita M and Meakin P 1988 *Phys. Rev.* A **37** 3645
Meakin P 1983 *Phys. Rev. Lett.* **51** 1119
—— 1984 *J. Chem. Phys.* **81** 4637
—— 1985 *J. Chem. Phys.* **83** 3645
—— 1988 *Phase Transition and Critical Phenomena* vol 12 ed C Domb and J L Lebowitz (Academic: London)
—— 1989 *Phys. Rev.* A **40** 2655
Meakin P and Jullien R 1985 *J. de Physique* **46** 1543
Meakin P, Vicsek T and Family F 1985 *Phys. Rev.* B **31** 564
Racz Z 1985*a* *Phys. Rev.* A **32** 1129
—— 1985*b* *Phys. Rev. Lett.* **55** 1707
Racz Z and Vicsek T 1983 *Phys. Rev. Lett.* **51** 2382
Salpeter E E 1977 *Ann. Rev. Astron. Astrophys.* **15** 267
Samsel R W and Perelson A S 1982 *Biophys. J.* **37** 493
Smoluchowski M V 1917 *Z. Phys. Chem.* **92** 129, 155
Spouge J L 1985 *J. Coll. Interface Sci.* **107** 38
Takayasu H 1989 *Phys. Rev. Lett.* **63** 2563
Takayasu H, Nishikawa I and Tasaki H 1988 *Phys. Rev.* A **37** 3110
Takayasu M and Takayasu H 1989 *Phys. Rev.* A **39** 4345
Thompson B R 1989 *J. Phys. A: Math. Gen.* **22** 879
van Dongen P G J 1987 *J. Phys. A: Math. Gen.* **20** 1889
Vicsek T, Meakin P and Family F 1985 *Phys. Rev.* A **32** 1122
White W H 1982 *J. Coll. Interface Sci.* **87** 204
Witten T and Sander L M 1981 *Phys. Rev. Lett.* **47** 1400
Ziff R M 1984 *Kinetics of Aggregation and Gelation* ed F Family and D P Landau (Amsterdam: Elsevier) p 191