Scaling in steady-state cluster-cluster aggregation

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The diffusion-limited cluster-cluster aggregation model is investigated under conditions which for long times lead to steady-state coagulation. Single particles are added to the system at a constant rate and the larger clusters appearing as a result of the aggregation process are removed according to various rules. Our results show that the dependence of the number of clusters, $N(t)$, on the feed rate κ in a unit volume and the time t can be well represented by a scaling form $N(t)\sim\kappa^{\alpha}f(\kappa^{\beta}t)$ with a scaling function $f(x)\sim x$ for $x\ll 1$ and $f(x)=1$ for $x\gg 1$. The exponents α and β are found to depend on the spatial dimension d, of the system, but within the statistical errors they always satisfy the relation $\alpha+\beta=1$ in accordance with the prediction of a generalized rate equation discussed by Rácz (see the companion paper). The values we have obtained for α and β are consistent in two and three dimensions with the corresponding results of the Smoluchowski equation approach but inconsistent in one dimension. This can be considered as an indication of the fact that the upper critical dimension for the kinetics of the diffusion-limited cluster-cluster aggregation model is 2.

I. INTRODUCTION

There has been a rapidly growing interest recently in the irreversible growth processes since they represent one of the most common phenomena in nature. Examples include nucleation, coagulation, flocculation, gelation, dendritic growth, polymerization, and aggregation processes which are related to various fields of science and technology.[1–3] A number of models have been proposed to study the geometrical properties of kinetically growing clusters, such as diffusion limited aggregation,[4] kinetic gelation,[5] and cluster-cluster aggregation.[6–8] It has been found that the clusters grown according to a nonequilibrium rule are typically fractals, having a fractal dimension[9] D less than the Euclidean dimension of the space in which the growth process takes place.

The diffusion-limited cluster-cluster aggregation model (CCA) introduced by Meakin[7] and Kolb *et al.*[8] is a particularly suitable model for studying multiparticle aggregation. Using this model it is possible to study not only the static geometrical properties of the clusters, but also the time evolution of the system in a class of coagulation phenomena in which the rate-limiting process is the diffusion of clusters (reorganization of the particles within the clusters is neglected in these models). In CCA the initially randomly distributed particles undertake a random walk and stick to each other on contact to form a cluster. The clusters continue to diffuse and by joining rigidly together form larger clusters which were shown to be fractals.[7,8] As the time proceeds the number of clusters of size s in a unit volume, $n_s(t)$, changes in a way which can be described in terms of dynamic scaling. Dynamic scaling for the cluster-size distribution in CCA for mass-independent diffusion coefficients was first proposed by Vicsek and Family,[10] while the case of mass-dependent diffusivity was treated by Kolb[11] and Meakin *et al.*[12]

Cluster-cluster aggregation in its original form describes a process with a permanent evolution in time as the number of clusters in the system is always decreasing. In this paper we modify the model to simulate an important process of both practical and theoretical interest, in which a steady-state cluster-size distribution develops in the system. This goal can be achieved by feeding single particles into the system and removing the larger clusters according to some rules. Steady-state conditions are very typical in many applied fields. For example, cars (as well as other sources of smoke) represent a permanent source of small smoke particles fed into the atmosphere. These smoke particles coagulate and form larger, heavier particles which gradually disappear from the atmosphere by sedimentation due to the gravitational force. In the stirred tank reactors for aerosols, often used for modeling chemical reactors in industry, an analogous process takes place but the particles are removed by letting them flow out from the chamber.[3,13]

We have simulated two different models of steady-state coagulation by feeding single particles into the system and by removing some of the larger clusters according to a given rule. At every unit time k particles are added at k different sites selected randomly. There are a few possi-

bilities for the removal of the clusters. We have considered two models. In model I, a cluster is discarded as soon as it becomes larger than a previously fixed number s_r. This is an extreme version of the situation in which the larger clusters leave the system with a higher probability. Another possibility—which is used in model II—is to remove the clusters with some rate r independent of the size of the clusters. Here the probability that a given cluster is removed during a unit time is proportional to r. In this way both the total number of clusters $N(t)$ and the number of particles $M(t)$ in the system of size L^d go to a constant value (N_∞ and M_∞) for long times.

Steady-state coagulation has been investigated by various approaches including experiments,[14,15] numerical methods,[16,17] and studies of a rate equation[18–20] for the cluster-size distribution usually called the Smoluchowski equation (SE). In the latter approach the mass distribution of diffusing clusters is determined from a system of differential equations which describe binary collisions with a constant rate K_{ij}, depending only on the size (mass) of the clusters. As an approximation to the true kinetics of the cluster-cluster aggregation process SE has been considered by a number of authors.[21–25] Since the spatial fluctuations are not accounted for in the Smoluchowski coagulation equation the applicability of this approach to the description of the time dependence in various aggregation models has been a subject of recent investigations. In general, because of its mean-field character SE is expected to give good or exact results above a certain critical dimension. In a three-dimensional simulation of the CCA model the collision frequencies were numerically determined and found to be time independent[24] in accordance with the assumption used in SE. It was also shown that SE is appropriate for describing the coagulation process in a two-dimensional aggregation model in which the diameter of the clusters is equal to unity independently of their mass (the clusters are infinite dimensional). However, in this model[23] a time dependent K_{ij} was needed for agreement with the one-dimensional simulation results.

The Smoluchowski equation was originally derived for describing the coagulation of compact (nonfractal) clusters. Its predictions for the case of fractal objects such as cluster-cluster aggregates and for low dimensions may considerably deviate from the true behavior of the diffusion-limited cluster-cluster aggregation process. Simulation of the steady-state aggregation process is particularly suitable for testing the region of validity of the Smoluchowski equation for CCA. Choosing the feed rate k as a small parameter it is possible to show[16,20,21] that in the SE approach (unless K_{ij} is time dependent), the number of clusters in the steady state N_∞ scales as $N_\infty \sim k^{1/2}$. From a study of the scaling properties of SE Rácz[21] showed that the characteristic time τ needed for relaxing to the final state scales as $\tau \sim k^{1/2}$, as well. In the case of a more general coagulation equation, $N_\infty \sim k^\alpha$ and $\tau \sim k^\beta$, with the exponents α and β satisfying the scaling relationship $\alpha+\beta=1$ independent of K_{ij}. Thus, in the steady-state coagulation there are two exponents which do not depend on the details of the aggregation process, while for the case of the ordinary coagulation kinetics the exponents obtained from the Smoluchowski equation depend on the particular (usually arbitrary) form of the reaction rate constant K_{ij}. Therefore, perhaps the best way to estimate the critical dimension for CCA kinetics and to investigate the effects of the fractal geometry of the clusters on the time dependence of the aggregation process is to calculate the exponents α and β from simulations and compare these results with the predictions of the SE.

The outline of the paper is as follows. In Sec. II we describe the simulations and present the results. The results are discussed in Sec. III. Finally, in Sec. IV we present our conclusions.

II. SIMULATIONS AND RESULTS

Our simulations were carried out in one to three dimensions on cells with linear size L and with periodic boundary conditions. Square and cubic lattices were used in two and three dimensions, respectively. The aggregation process is started with M_0 randomly distributed particles at the sites of the cells. The particles and the clusters undergo a random walk and stick rigidly together on contact to form new clusters. The time is measured by the number of attempted moves divided by the total number of clusters $N(t)$ in the system. In general, the diffusion constant of a cluster may depend on the size of the cluster s (where s is the number of particles in the cluster). We used a size- (mass-) dependent cluster diffusivity of the form $D_s = D_0 s^\gamma$. When making an attempt to move, a cluster is shifted by one lattice unit only with a probability proportional to its mobility, but the time is incremented independently of whether the move has or has not been made.

In the following we shall concentrate on the dependence of the approach to the steady state and the steady state itself on the feed rate k. In order to do this we calculate for various k the quantities $N(t)$, $M(t)$, and $N_s(t)$, where $N_s(t)$ is the number of clusters of size s in the system. After a sufficiently long time these quantities become (on the average) time independent and their values N_∞, M_∞, and $N_{s,\infty}$ can be used to describe the steady state. We call the characteristic time needed for approaching the final state from a low-density initial state the relaxation time (τ). In most of the simulations the number of clusters (or particles) exhibits a damping oscillatory behavior before it assumes its steady-state value. One way of defining the relaxation time is to identify τ with the time at which the first maximum or some similar feature such as the first minimum or second maximum appears in $N(t)$.

In order to obtain more accurate results we calculate various kinds of averages. First, the same quantity is calculated from a number of runs (typically this number is about 20). Second, a time average is taken in the steady state. Finally, for large cluster sizes the results for a small range of cluster sizes are averaged to obtain a better estimate of $N_{s,\infty}$.

In one dimension the cluster-cluster aggregation process has been simulated on a line of length $L=131\,072$ (2^{17}) or $L=65\,536$ (2^{16}) lattice units. In the low-density limit the size of the clusters is always much less than the distance between them; therefore, the asymptotic behavior of the kinetics of CCA in one dimension is expected to be the same as in the simple model of Kang and Redner.[23] In the latter model when two clusters of masses i and j meet,

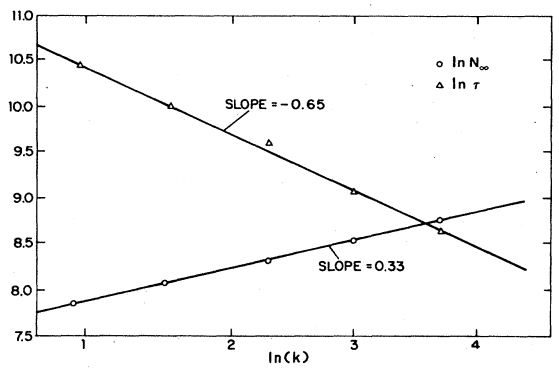

FIG. 1. The number of clusters in the steady state N_∞ and the time τ required to reach the first maximum in the time-dependent number of clusters vs the particle feed rate k. In these simulations carried out on a linear chain of length $L=131\,072$ (2^{17}), k was measured in particles/(time unit) and the time was in units of attempted moves per cluster. The maximum cluster size was s_r (clusters containing more than 250 particles were removed from the system as soon as they were formed). The straight lines on this log-long plot indicate scaling of the quantities N_∞ and τ.

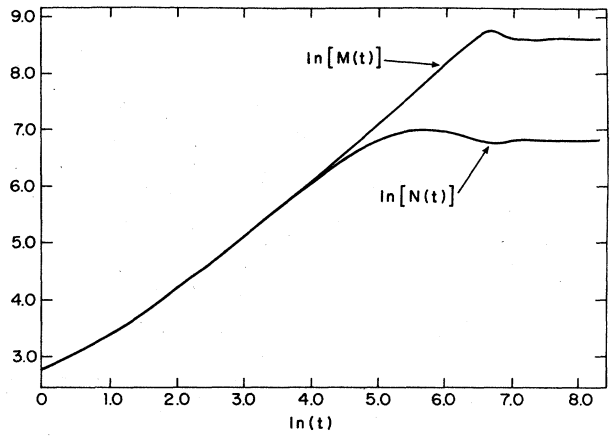

FIG. 3. Time dependence of the total number of clusters, $N(t)$, and total number of particles in the system, $M(t)$, obtained from simulations of cluster-cluster aggregation on 400^2 lattices with a feed rate of 8 particles/(time unit). Clusters with a size greater than $s_r=100$ were removed.

they coalesce into a single particle (one lattice site) of mass $i+j$. Using this assumption we have obtained data for the steady-state number of particles N_∞ and for the relaxation time τ as a function of k. The results are displayed in Fig. 1 for model I with $s_r=250$ and $\gamma=0$. The distribution of the cluster sizes after a long time is shown in Fig. 2.

The two-dimensional simulations were carried out on 400^2 lattices. Figure 3 shows the behavior of $N(t)$ and $M(t)$ as a function of time. In these simulations all clusters larger than $s_r=100$ were removed from the system (model I) and a feed rate $k=8$/(time unit), a size-independent diffusion constant, and a low initial number of particles $M_0=16$ (initial density $\rho=0.0001$) were used. In Fig. 4(a) we display data obtained for the number of clusters (N_∞) in the steady state and the relaxation time τ for various feed rates k. The straight lines on this log-log plot indicate scaling of N_∞ and τ as a function of k.

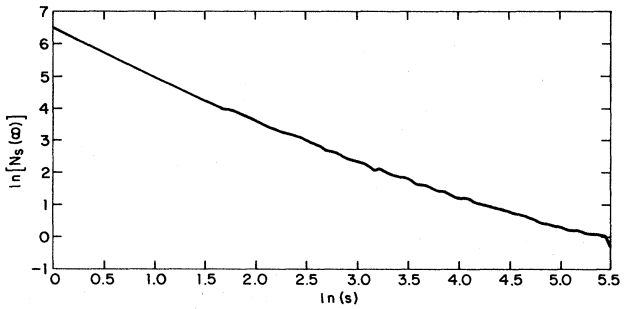

FIG. 2. The steady-state cluster-size distribution obtained from a one-dimensional simulation. The lattice size was $L=65\,536$ (2^{16}), the particle feed rate was 5 particles per time unit, and the maximum allowed cluster size was equal to $s_r=250$. The results shown in Figs. 1 and 2 were obtained with mass-independent diffusion coefficients.

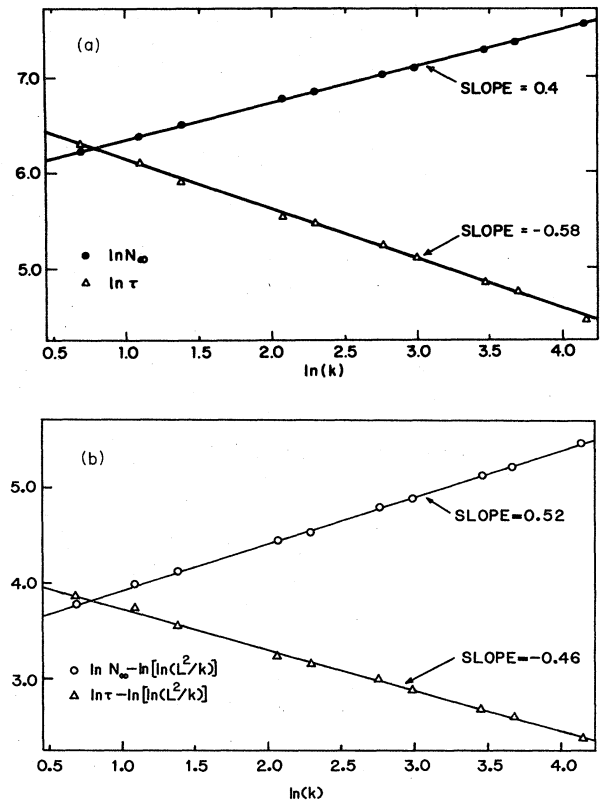

FIG. 4 The total number of clusters in the steady state N_∞ and the time τ required to reach the first maximum in $N(t)$ (shown in Fig. 3) vs the particle feed rate k (a). The scaling of N_∞ and τ as a function of k is indicated by the straight lines drawn through the data on this log-log plot. Assuming the existence of a logarithmic correction of the form $N_\infty \sim k^\alpha \ln(L^2/k)$ one is able to determine the exponent α from the plot $\ln N(t) - \ln[\ln(L^2/k)]$ vs $\ln(k)$ (b).

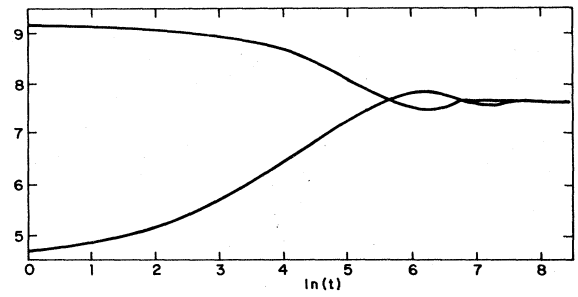

FIG. 5. Time dependence of the total number of clusters $N(t)$ obtained from three-dimensional simulations of cluster-cluster aggregation on 128^3 with particle feed rate of 10 particles/(time unit). The maximum allowed cluster size was $s_r=250$ and the initial particle concentrations were 10000 and 100 particles.

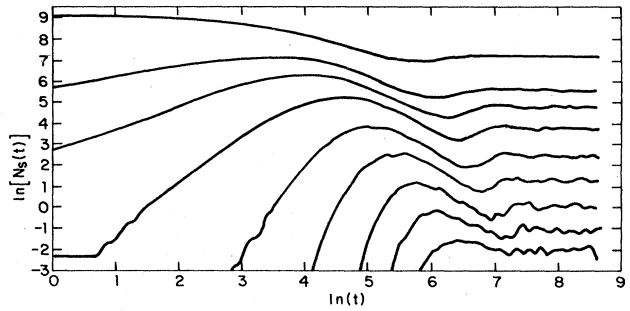

FIG. 7. The time dependence of the number of clusters of size s, $N_s(t)$, for various cluster sizes (1, 2, 3, 5, 9—10, 19—20, 38—40, 75—80, and 150—160) obtained during a three-dimensional simulation of cluster-cluster aggregation. The starting concentration was 10000 particles, 10 particles were added to the system at every time unit, and s_r was equal to 250.

However, if $d=2$ is the upper critical dimension for the kinetics of the CCA, the logarithmic corrections have to be taken into account as well. Assuming that $N_\infty \sim k^\alpha \ln(L^2/k)$ we are able to determine the exponent α by plotting $\ln N_\infty - \ln[\ln(L^2/k)]$ versus $\ln(k)$ as it is presented in Fig. 4(b). We also calculated these quantities for the case where the diffusivity of the clusters depends on their size s as $D_s \sim 1/s$. Our results show that the exponents describing the power-law behavior of N_∞ and τ are insensitive to the change in the diffusivity of the clusters.

Since most of the practically interesting processes which are analogous to the steady-state cluster-cluster aggregation occur in three dimensions we have carried out more extensive simulations on 128^3 lattices. In the first series of simulations we studied model I. In order to see the effects of k, r, s_r, and M_0 on the results, runs with various values of these parameters were made. Figure 5 shows the time dependence of $N(t)$ for two initial values of the number of particles $M_0=100$ and $M_0=10000$, with $k=10$ and $s_r=250$. The behavior of N_∞ and τ as a function of the feed rate k is presented in Fig. 6. In these simulations a low initial value $M_0=100$ was used for the number of particles. The data show that N_∞ and τ scale with k in three dimensions, too. We have also calculated the dynamic cluster-size distribution $N_s(t)$. Figure 7 shows the time dependence of $N_s(t)$ for a few selected cluster sizes and for $M_0=10000$, $k=10$/(time unit) and $s_r=250$. The steady-state distribution for this case is presented in Fig. 8. Additional simulations show that increasing s_r to 500 or to 1000 does not have a significant effect on N_∞ and the only important consequence of this change is that the mean cluster size in the steady state

$$S(t=\infty)=\sum_s s^2 N_{s,\infty} / \sum_s (sN_{s,\infty})$$

increases.

Next we investigated the second version of the steady-state models. In model II, single particles were added to the system every unit time as before, but the probability of removing any of the clusters during a unit time was equal to a small constant r. In this model the dependence of the number of clusters does not show a maximum but there is a maximum in the plot of the mean cluster size (Fig. 9).

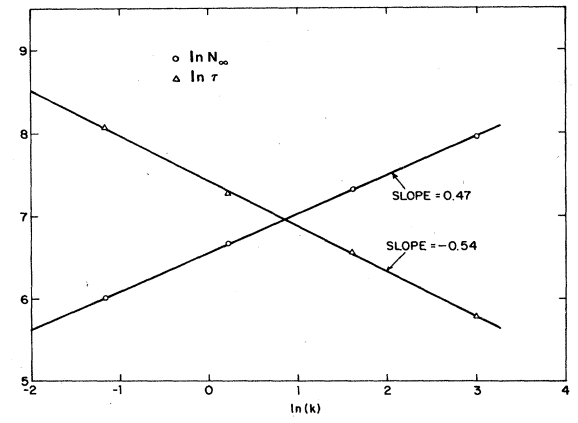

FIG. 6. This figure shows how the total number of clusters in the steady state N_∞ and the time τ needed to reach the first maximum in $N(t)$ depend on the particle feed rate k in the simulations of coagulation on 128^3 lattices.

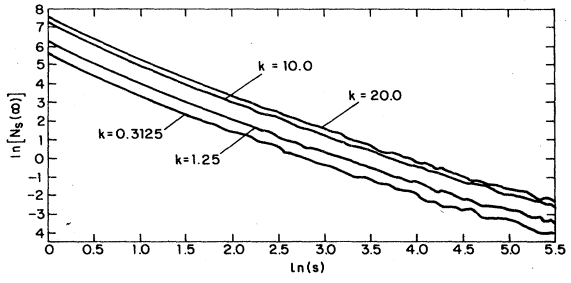

FIG. 8. The steady-state cluster-size distribution under the conditions used to obtain Fig. 7. Results are also shown for the feed rates 0.3125, 1.25, and 20 particles/(time unit).

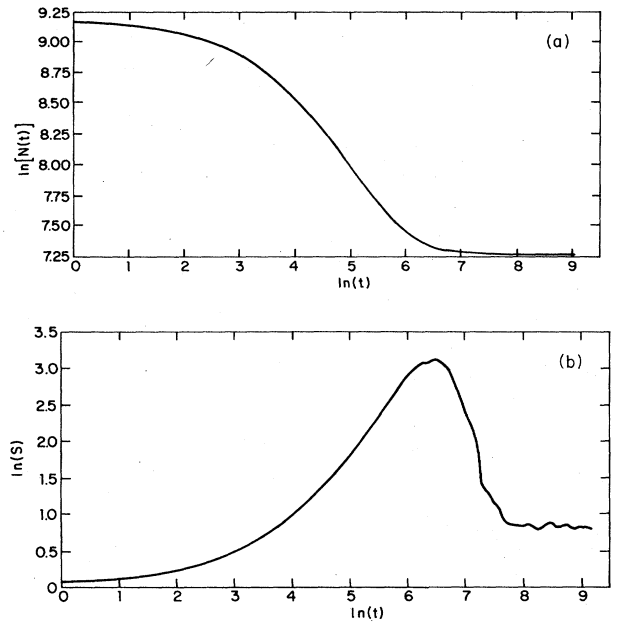

FIG. 9. This figure shows some typical results obtained from a simulation carried out using model II. The simulation was started with 10000 particles on a 128^3 lattice and additional particles were fed in at a rate of 10 particles/(time unit). The rate constant for removal of clusters of all sizes was equal to $r=0.005$/(time unit). (a) shows the time dependence of the number of clusters, $N(t)$, and in (b) the mean cluster size as a function of time, $S(t)$, is displayed.

The cluster-size distribution after a long time for a constant removal rate $r=0.005$ and for various diffusivities ($\gamma=0.0$, $-1/D$, and -1.0) is presented in Fig. 10. This figure shows that the cluster-size distribution follows a power law with a likely exponential cutoff for the large values of s. The value of the exponent describing this algebraic decay depends only slightly on γ.

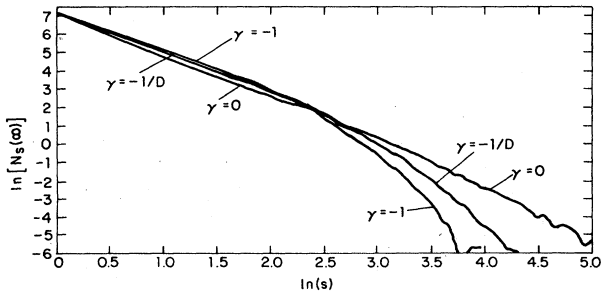

FIG. 10. Steady-state cluster-size distributions from three-dimensional simulations with a particle feed rate of 10 particles/(time unit) and with a constant removal rate of $r=0.005$/(time unit) (model II). Results are shown for several values of the exponent γ which determines the diffusivity of the clusters of size s, D_s, through the expression $D_s \sim s^{\gamma}$. The maximum allowed cluster size in these simulations was 250 particles.

III. DISCUSSION

A characteristic feature of the approach to the steady state is that the moments of the dynamic cluster-size distribution $N_s(t)$ show an oscillatory behavior in most of the cases (Figs. 3 and 5). This behavior can qualitatively be explained as follows. In the steady-state models discussed in this paper there are three processes affecting the number of clusters $N(t)$ in the system. The addition of particles with a given feed rate increases $N(t)$, while the coagulation (sticking together) and the removal of the clusters decreases it. As the clusters grow and the distance among them becomes smaller [with increasing $N(t)$ and cluster radius], the rate of coagulation increases and compensates the feed rate. At this moment the number of clusters starts decreasing as in the ordinary CCA. This decline in $N(t)$, however, does not continue forever, since as soon as clusters larger than s_r are formed they are removed from the system. In this way the clusters which were the most effective in "capturing" other clusters are eliminated and the tendency changes. In model II this process is smoothed out, but an extremum still can be seen on the plots of the mean cluster size (Fig. 9).

Our results for the steady-state value of the number of clusters N_∞ and for the relaxation time τ indicate scaling of these quantities as a function of the feed rate k. Figure 11 shows how the steady-state cluster-size distribution $N_s(\infty)$ can be scaled as a function of k using a simple scaling form $N_s(\infty) \sim k^{\alpha} g(s)$. In the case when the initial density of the particles is very small our data for $N(t)$ given in Fig. 12(a) show that the total number of particles in a unit volume at time t, $n(t)$, can be well represented by the following scaling form:

$$n(t) \sim \kappa^{\alpha} f(\kappa^{\beta} t) , \tag{1}$$

where κ is the feed rate in a unit volume and $f(x)$ is a scaling function with $f(x) \sim x$ for $x \ll 1$ and $f(x)=1$ for $x \gg 1$. This behavior is demonstrated in Fig. 12(b) where the $N(t)$ curves obtained in the three-dimensional simulations for various feed rates are scaled into one universal function. The actual shape of $f(x)$ may depend on the parameters γ, s_r, or r but for a fixed set of these numbers $N(t)$ can be expressed through the scaling form (1).

The values of the exponents α and β can be determined from the slopes of the straight lines drawn through the data on the log-log plots of N_∞ and τ versus k. The

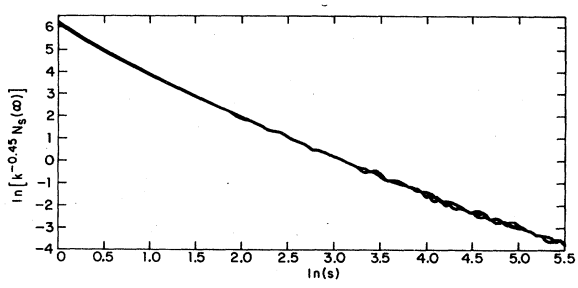

FIG. 11. The steady-state cluster size distributions shown in Fig. 8 for various feed rates k scaled onto a single universal curve.

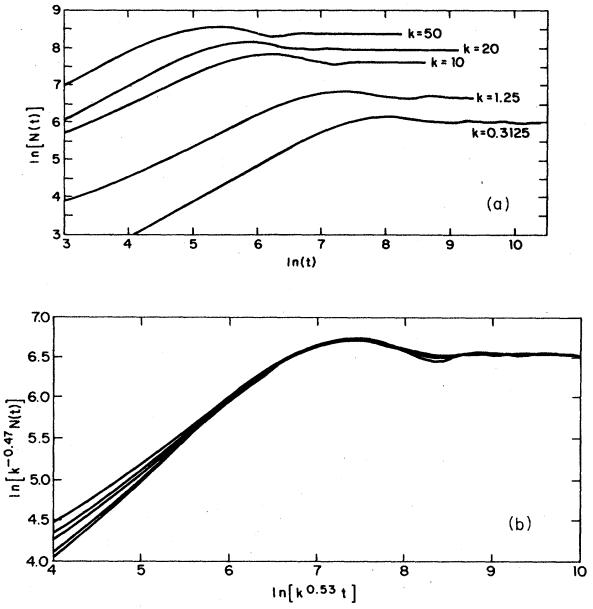

FIG. 12. This figure shows the time dependence of the total number of clusters obtained from three-dimensional simulations of cluster-cluster aggregation with feed rates of 0.3125, 1.25, 10, 20, and 50 particles/(time unit) and with a maximum allowed cluster size $s_r=250$. (a) shows the data before scaling and (b) shows how the scaling form $N(t)\sim k^{\alpha}f(k^{1-\alpha}t)$ can be used to scale curves in (a) onto a single universal curve. The deviations from scaling apparent on the left-hand side of (b) are a result of the fact that instead of a zero initial particle concentration ($M_0=0$), a small finite initial concentration was used in these simulations.

numbers we obtained for α and β depend on the dimension of the space in which the cluster-cluster aggregation takes place but they seem to be insensitive to the mass dependence of the diffusivity or to the parameter s_r. In one dimension we found that $\alpha=0.33\pm0.02$ and $\beta=0.65\pm0.03$. The two-dimensional simulations gave $\alpha=0.40\pm0.04$ and $\beta=0.58\pm0.05$ without, and $\alpha=0.52\pm0.04$ and $\beta=0.46\pm0.05$ with logarithmic corrections taken into account, while in three dimensions $\alpha=0.47\pm0.05$ and $\beta=0.54\pm0.05$ were obtained.

In addition to the simulations the one-dimensional case allows an approximate treatment using a rate equation for the number of clusters

$$\frac{dN(t)}{dt}=k-bN^3(t)-F(k,s_r,t)\ , \tag{2}$$

where k is the feed rate, b is a constant, and the term $-F(k,s_r,t)<k/s_r$ describes the removal of clusters as it is done in model I. The term $N^3(t)$ is included because of the following consideration. The rate of change of $N(t)$ due to coagulation is proportional to the number of clusters itself and to the average collision frequency of the clusters ν. In one dimension because of the diffusional motion this frequency is inversely proportional to the square of the average distance R between the clusters. On the other hand, $R=L/N(t)$; therefore, $\nu\sim N^2(t)/L^2$, where L is the length of the chain along which the clusters diffuse. Although the above arguments are not rigorous, they are quite plausible, and therefore Eq. (2) is expected to give the exact asymptotic behavior of $N(t)$. For $k=0$ and $s_r\to\infty$ it gives $N(t)\sim t^{1/2}$ in agreement with Kang and Redner's simulations and the Smoluchowski equation results.[23]

From (2) it can be seen that for $s_r\to\infty$ in the steady state the number of clusters is equal to $(k/b)^{1/3}$; therefore, N_∞ scales with the feed rate with an exponent $\alpha=\frac{1}{3}$. In order to get an expression for the relaxation time we integrate Eq. (2) for $s_r\gg1$ and obtain

$$-t=\frac{1}{6ba^{2/3}}\ln\frac{[N(t)-a^{1/3}]^2}{N^2(t)+N(t)a^{1/3}+a^{2/3}}+\frac{1}{3ba^{2/3}}\tan^{-1}\frac{2N(t)+a^{1/3}}{3a^{1/3}}+C\ , \tag{3}$$

where $a=k/b$ and C is a constant. Keeping only the term which becomes singular as $N(t)\to a^{1/3}$ we get from (3)

$$N(t)\simeq a^{\alpha}-\lambda e^{-t/\tau}\ , \tag{4}$$

with the characteristic time $t=k^{-\beta}/3b^{1/3}$ and with $\alpha=\frac{1}{3}$ and $\beta=\frac{2}{3}$. In Eq. (4) λ is a constant depending on the initial conditions. The values $\alpha=\frac{1}{3}$ and $\beta=\frac{2}{3}$ satisfy the equality $\alpha+\beta=1$ and are in good agreement with the simulation results. For model II similar considerations result in the same values for the exponents α and β.

Having determined α and β in one to three dimensions we are able to discuss the relevance of the Smoluchowski equation to the cluster-cluster aggregation under steady-state conditions. One of the notable features of the numbers obtained for the exponents α and β is that their sum is approximately equal to one $\alpha+\beta\simeq1.0$ in all dimensions we investigated. As it was shown by Rácz[21] a generalized version of the Smoluchowski equation for the cluster numbers $N_s(t)$ leads to the same result. On the other hand, from the scaling properties of the original SE, it follows[16,20,21] that α and β should be equal to $\frac{1}{2}$ in all dimensions. This value is in clear contradiction with our simulation results in one dimension, while it is in a reasonable agreement with the results obtained for the two- and three-dimensional cases.

IV. CONCLUSIONS

We have presented results of computer simulations studies of diffusion-limited cluster-cluster aggregation under conditions leading to steady-state coagulation. The study of steady-state models is particularly suitable for investigating the applicability of the Smoluchowski equation to coagulation processes. The exponents α and β describing the asymptotic dependence of the number of clusters and the relaxation time do not depend on the particular form of the rate constant K_{ij}. This is in contrast with the non-steady-state models where the value of K_{ij} has a strong effect on the behavior of the systems. Our results indicate scaling as a function of the feed rate with exponents which are not consistent with the mean-field approach (SE) below two dimensions. They are in accor-

dance, however, with the theoretical prediction $\alpha+\beta=1$ for all the cases we have considered. It has recently been shown[23] that the upper critical dimension d_c for the kinetics of the model of single-site clusters is equal to 2. The α and β values we obtained for the two-dimensional case are consistent with the mean-field result only if we assume that $d_c=2$ for the kinetics of the CCA model and take the logarithmic corrections into account.

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