Reversible aggregation in an off-lattice particle-coalescence model: Dynamical and steady-state scaling behavior

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(Received 3 February 1992)

We have performed numerical simulations in an off-lattice version of the reversible particle-coalescence model. We have analyzed both two- and three-dimensional systems for several values of the coagulation and fragmentation rate constants. The numerical data support recent conjectures by Sorensen, Zhang, and Taylor [Phys. Rev. Lett. **59**, 363 (1987)] on dynamical scaling behavior except at very early times, when we see crossover effects coming from the aggregation-dominated regime.

PACS number(s): 82.20.Wt, 05.40.+j

I. INTRODUCTION

A considerable amount of work has recently been devoted to understanding the intrinsic mechanism of the irreversible aggregation process of small particles to form large clusters. The mechanism is of interest in many areas of physics, chemistry, biology, and astronomy [1–3]. Besides, the structures generated in this way, because of their unique properties, are of considerable importance in many basic and applied problems in polymerization, gelation, and colloidal science [4–8]. A general feature is that, despite the broad origin of these phenomena, the resulting structures appear in many cases to be scale invariant and can be generally treated as fractals [9–12]. As a consequence, the mean cluster size $S(t)$ is found to increase with time algebraically as $S(t) \sim t^z$ and the cluster-size distribution $n_s(t)$, defined as the number of clusters of size s per unit volume at time t, obeys a scaling form in the following way:

$$n_s(t) = s^{-2}\phi_0(u), \tag{1}$$

where $u = s/S(t)$. The above results are well established in experimental [13], computer [14], and theoretical [15] studies.

A more general and realistic description of the aggregation process should include the possibility that aggregates can break into small pieces, the so-called reversible aggregation process [16–22]. The global process can be schematically represented by the following reaction mechanism:

$$A_i + A_j \underset{F(i,j)}{\overset{K(i,j)}{\rightleftharpoons}} A_{i+j},$$

where A_i denotes a cluster containing i elementary units, and $K(i,j)$ and $F(i,j)$ are the forward and reverse rate coefficients representing the coagulation and fragmentation processes, respectively.

The mean-field rate equation, which describes the evolution of the cluster-size distribution under the assumption of binary collisions, is given by the generalized Smoluchowski equation (to be referred as GS) [23–25]:

$$\frac{dn_s}{dt} = \frac{1}{2}\sum_{i+j=s}[K(i,j)n_i n_j - F(i,j)n_s] - \sum_{j=1}^{\infty}[K(s,j)n_s n_j - F(s,j)n_{s+j}]. \tag{2}$$

This equation gives the time variation of the s-mer concentration, $n_s(t)$, in terms of gains and losses due to different reactions in the sample. $K(i,j)$ is the coagulation kernel, describing the aggregation reaction rate of an i-mer with a j-mer. This kernel contains the i and j dependence of the meeting probability of an i-mer and a j-mer, including effects such as the mass dependence of the collision cross section and the diffusion constant. $F(i,j)$ is the fragmentation kernel describing the breakup of an $(i+j)$-mer into an i-mer and a j-mer.

The competition between coagulation and fragmentation processes may lead to a final steady-state configuration. There the mean cluster size $S(\kappa, t=\infty)$ is expected to be determined by the breakup constant κ, measuring the relative strength of the rate constants for fragmentation and coagulation reactions. For small κ or for sufficiently large mean cluster size, $S(\kappa, t=\infty)$ decreases according to a power law: $S(\kappa, t=\infty) \sim \kappa^{-y}$ [17]. By assuming that the scaling ansatz, known to be valid for pure aggregation processes, also holds in the steady state, Family, Meakin, and Deutch [17] concluded that $n_s(\kappa,\infty) = s^{-2}\phi(u)$ with $u = s/S(\kappa,\infty)$. Later on, Sorensen, Zhang, and Taylor [18] (to be referred as SZT) extended the same scaling relation to all times. They also derived expressions for the mean cluster size at equilibrium and the characteristic time to reach equilibrium in terms of the strengths of the kernels. In their derivation

they assumed certain parameters to be constants. This fact was criticized by Vigil and Ziff [20], who showed that these parameters depend on the order of the moment used in the derivation of the time derivative for the mean cluster size. Also, Meakin and Ernst [19] developed the important result that the scaling form for the cluster size distribution is different in the early stage of the evolution, which is in the aggregation dominated regime, from the distribution in the later stages of evolution where the system approaches equilibrium. They assumed the existence of a characteristic time separating the evolving and equilibrium regimes $\tau(\kappa) \sim \kappa^{-x}$ for $\kappa \to 0$ where for $t \gg \tau(\kappa)$ the system had reached the steady state and for $t \ll \tau(\kappa)$ irreversible aggregation dominates. In the early time regime the characteristic mean cluster size was supposed to follow the scaling form $S(\kappa,t) \sim \kappa^{-y}\psi(T)$ with $T = t/\tau(\kappa)$. To match the behavior of irreversible aggregation as $\kappa \to 0$ the function ψ had to be of the form $\psi(T) \sim T^z$, for $T \to 0$, with $z = y/x$, so that $S(\kappa \to 0, t)$ was independent of κ. Again, to match the steady state $\psi(T)$ must tend to a constant as $T \to \infty$, so $\psi(\infty) = 1$. Finally the cluster size distribution $n_s(\kappa,t)$, depending on three arguments, approaches in the scaling limit a form dependent only on two arguments:

$$n_s(\kappa,t) = s^{-2}\phi(u,T) \tag{3}$$

with $u = s/S(\kappa,t)$. In order to match irreversible and steady-state scaling form it was imposed that $\phi(u,0) = \phi_0(u)$ and $\phi(u,\infty) = \phi(u)$. Therefore the simple scaling assumption used by SZT was not supported and their results must be questioned. However, the characteristic time to approach equilibrium defined by Meakin and Ernst [19] and the stability criterion found by Vigil and Ziff [21] were correctly obtained by SZT [18]. Recently Elminyawi, Gangopadhyay, and Sorensen [22] have solved Eq. (2) numerically in order to investigate the validity of the predictions of SZT. They compared the numerical values obtained for the mean cluster size at equilibrium and the time taken to reach equilibrium to those obtained from the theoretical expressions of SZT. The good agreement obtained for the mean cluster size was in contrast with the systematic differences found in the characteristic time. It was not clear whether the discrepancies resulted from the approximations introduced by them in the scaling function for the size distribution or from the approximations in the work of SZT criticized by Vigil and Ziff [20]. It seems that the SZT theory yields an asymptotic solution of the GS equation, which, however, cannot give accurate results in the intermediate time stage. However, their asymptotic scaling predictions in which most of the parameters are absorbed in the rescaled variables can be tested against computer simulation of simplified models.

Since the numerical simulations carried out before have all been restricted to very simple lattice models, the validity of the description of the evolution process in terms of scaling laws should be tested in a more realistic model, based on off-lattice simulations, where the locations and movements of the particles are not constrained to a regular lattice. This could be useful in determining the range of validity of the mean-field equations and in evaluating how severe are the approximations introduced by SZT in extending the same scaling law for the size distribution used in the steady-state limit to all time in the aggregation-fragmentation process. We present in this paper the results of a numerical study of the reversible aggregation process in an off-lattice coalescence model in two- and three-dimensional systems. We have paid attention to both dynamical and steady-state scaling behaviors. We have computed the mean cluster size and the size distribution at equilibrium. We have tested the scaling predictions, which are in agreement with our computer simulations. We have also computed the time evolution of the mean size, which agrees reasonably well with the theoretical predictions made by SZT.

The rest of the paper is organized as follows. In Sec. II we present a theoretical review of the reversible aggregation process. In Sec. III we describe the numerical procedure. In Sec. IV A we compute the time evolution of the mean cluster size from the aggregation-dominated regime to equilibrium. Section IV B deals with the steady-state limit for the cluster-size distribution and the mean size. Section V concludes with a discussion of the results.

II. THEORETICAL REVIEW

SZT started with the generalized Smoluchowski equation, Eq. (2), to study the mean-cluster-size evolution. There the coagulation and fragmentation kernels were represented by $K(i,j) = \kappa_c\psi(i,j)$ and $F(i,j) = \kappa_f\Phi(i,j)$, with $\psi(1,1) = \Phi(1,1) = 1$ and κ_c and κ_f being the coagulation and breakup constants, respectively. They assumed a cluster-size distribution scaling behavior as

$$n_s(\kappa,t) = M_1 S(\kappa,t)^{-2}\varphi(u), \tag{4}$$

with $u = s/S(\kappa,t)$. M_1 is the first moment in the size distribution and represents the total number of particles in the system, which is a constant. The moments are calculated in the usual way,

$$M_i = \sum_{s=1}^{\infty} s^i n_s(\kappa,t), \tag{5}$$

and the mean cluster size is defined as

$$S(\kappa,t) = M_2/M_1. \tag{6}$$

The kernels were assumed to be homogeneous functions of their arguments, so that $K(ai,aj) = a^\lambda K(i,j)$ and $F(ai,aj) = a^\alpha F(i,j)$. By writing the equation for the time derivative of the second moment, one finally finds a differential equation for $S(\kappa,t)$:

$$\frac{d}{dt}S(\kappa,t) = M_1 A\kappa_c S^\lambda - B\kappa_f S^{\alpha+2}, \tag{7}$$

where

$$A = \int_0^\infty dx \int_0^\infty dy\, xy\psi(x,y)\varphi(x)\varphi(y),$$
$$B = \int_0^\infty dx \int_0^\infty dy\, xy\Phi(x,y)\varphi(x+y). \tag{8}$$

One of the key points argued by Vigil and Ziff [20] was that A and B were dependent on the order of the moment used in deriving Eq. (7). For our purpose we consider A and B to be constants here and will later discuss the validity of this approximation.

Rewriting Eq. (7) in terms of the reduced variables

$$\hat{s} = \frac{S(\kappa,t)}{S(\kappa,\infty)}, \tag{9}$$

$$\hat{t} = \frac{t}{\tau}, \tag{10}$$

one obtains the scaled equation,

$$\frac{d}{d\hat{t}}\hat{s} = \hat{s}^{\lambda} - \hat{s}^{\alpha+2}. \tag{11}$$

The mean cluster size at equilibrium is found to be

$$S(\kappa,\infty) = \left(\frac{M_1 A \kappa_c}{B\kappa_f}\right)^{y}, \tag{12}$$

and the characteristic time to approach equilibrium,

$$\tau = (M_1 A \kappa_c)^{-(\alpha+1)y} (B\kappa_f)^{(\lambda-1)y}, \tag{13}$$

where

$$y = (\alpha - \lambda + 2)^{-1}. \tag{14}$$

The characteristic time separating the evolving and equilibrium regimes introduced by Meakin and Ernst [19] scaled as $\tau(\kappa) \sim \kappa^{-x}$ with $\kappa = \kappa_f/\kappa_c$, while SZT's result [Eq. (13)] shows that the characteristic time scales separately with κ_f and κ_c, with different exponents for the coagulation and breakup constants. For the case $\kappa_c = 1$, which will be considered later, SZT's results reduce to the Meakin and Ernst hypothesis and the exponent x is determined as $x = (1-\lambda)y$.

III. NUMERICAL MODEL

In order to test the scaling predictions for the cluster size distribution and the time evolution of the mean cluster size, we have carried out extensive numerical simulations in two- and three-dimensional off-lattice systems. We have considered a hard-sphere model for the particles enclosed in a cell or cube of dimensions $L \times L \times L$ (square of size $L \times L$ in two dimensions), where L is equal to 100 particle diameters. The diameter is taken to be the unit length. Our simulation starts by randomly placing a number of particles of unit mass in the cell in such a way that we cover 10% of the total volume (surface) available (that is, there are nearly 200 000 particles in the system in the three-dimensional case). The initial condition preserves the excluded-volume criterion, so that the particle placement is rejected if it overlaps with any of the remaining particles. We choose at random one of the clusters, initially identified with particles, and move it by a distance equal to its diameter in a randomly chosen direction. In this movement it may interact with another particle in the sense that the distance between the centers of these particles can become less than one diameter. In that case, they aggregate and form a cluster. Time is measured in units of *Monte Carlo time steps per cluster.*

In order to avoid the complexity arising from the geometrical structure of the clusters, which should be reflected in the functional form of the reaction kernels, we employ the so-called particle coalescence model [14]. In this model, the clusters are defined to be single particles with the same size as the initial ones. So, when two clusters of masses i and j meet, they coalesce into a heavier cluster of the same diameter and mass $i+j$ at a rate proportional to the reaction kernel $K(i,j)$. Since there is no cluster geometry, the functional form of the reaction kernel can be specified exactly. In our simulation we have assumed a mass-independent sticking probability, so that the aggregation kernel $\psi(i,j)$ is constant. This gives $\lambda = 0$. Furthermore, we consider $\kappa_c = 1$ (so that $\kappa = \kappa_f$), which yields simply $K(i,j) = 1$. Therefore Eq. (14) becomes

$$y = (\alpha + 2)^{-1}. \tag{15}$$

The probability of choosing a cluster of mass $s = i + j$ is given by $n_s(\kappa,t)$, and any of the $s-1$ bonds can be broken with equal probability. Then the probability of breaking one of the $s-1$ bonds of a cluster of size s can be written as

$$n_s(\kappa,t)\frac{1}{s-1}p(s), \tag{16}$$

where $p(s)$ is considered to have the following functional form:

$$p(s) = \kappa_f (s-1)\, s^{\alpha}, \tag{17}$$

so that $F(i,j) = \kappa_f \Phi(i,j) = \kappa_f (i+j)^{\alpha}$. Therefore the constants α and κ are the adjustable parameters in the model. In the simulation, a cluster is picked at random and an attempt is made to either break it or to move it also in a random fashion. If we attempt to break a cluster of size $s = i + j$, it will break with a probability given by $p(s)$. The new position of one of the two fragments will remain at the same location of the original cluster and the other one is placed at a random location inside the cell, preserving the excluded-volume criterion. This situation corresponds to the so-called *uncorrelated fission*: the resulting fragments are placed at random, uncorrelated positions, in the spirit of the mean-field theory. This choice of uncorrelated fission, coupled with the fact that the resulting geometry of clusters is neglected, make it plausible that the process is described by the GS equation, with single choices for the fragmentation and aggregation kernels.

In a general computer simulation, where long-range forces are present, each cluster A_i can potentially interact with any of the remaining aggregates. However, for short-range interactions, a significant amount of computer time, used in determining which pairs of clusters are close enough to interact, can be reduced. In order to achieve this, one should be able to efficiently compute the subset of clusters with which A_i interacts. For this reason we have implemented a link-cell (LC) method

[26–28]. In this scheme the simulation cell is broken into N_c smaller subcells. Then we assign to each subcell a list of clusters belonging to it. If the edge length of each cell L_c exceeds the diameter of the particles, then all interacting pairs are located within the same cell or one of the 26 adjoining cells (8 in two dimensions). Therefore the time to find all pairs of clusters that interact scales with the actual number of clusters, say N, instead of N^2 for the standard method. For each pair of values (κ, α) the maximum number of subcells N_c is chosen in order to minimize the CPU time.

IV. RESULTS AND DISCUSSION

A. Dynamical evolution

In order to investigate the validity of Eq. (11) and therefore the approximation introduced by SZT, we have solved Eq. (11) analytically for $\alpha = 1$ and numerically for $\alpha = -\frac{1}{2}$, with the initial condition for the mean cluster size equal to $\hat{s}(t = 0) = 1/S(\kappa, \infty)$. Then we have computed the mean cluster size $S(\kappa, t)$, the cluster-size distribution $n_s(\kappa, t)$, and the total number of clusters $n(\kappa, t) = M_0$ every 5×10^4 Monte Carlo time steps from $t = 0$ up to the steady state. The results are averaged over ten different initial conditions. In Figs. 1 and 2 we compare the data obtained in our numerical simulation with the analytical solution of Eq. (11) for $\lambda = 0$. We define a new reduced variable $\hat{n} = n(\kappa, t)/S(\kappa, \infty)$ and plot the analytical and numerical values for the reduced variables $\hat{s}$ versus $\hat{t}$, $\hat{n}$ versus $\hat{t}$, and $\hat{s}\hat{n}$ versus $\hat{t}$ on a logarithmic scale. In order to obtain the time rescaling parameter τ, we rewrite Eq. (13) in the form $\tau = (M_1 A \kappa_c)^{-1} S(\kappa, \infty)^{1-\lambda}$. In our case $\lambda = 0$ and $\kappa_c = 1$, so that $\tau = (M_1 A)^{-1} S(\kappa, \infty)$. Therefore $\tau = \frac{S(\kappa,\infty)}{M_1 A}$, where $M_1 A$ is an adjustable parameter in the model, which must be independent of α. In practice we have varied in every case the value of A to get the best agreement between theory and simulation. The values of A obtained in this manner for different values of α are constant within the numerical accuracy (for instance, $M_1 A = 0.67$ for $\alpha = 1$ and $M_1 A = 0.66$ for $\alpha = -\frac{1}{2}$). In Figs. 1 and 2, we plot the results for a three-dimensional system with a breakup constant $\kappa = 5 \times 10^{-4}$ and $\alpha = -\frac{1}{2}$ and $\alpha = 1$, respectively. We observe that the numerical data are in good agreement with the theoretical predictions and, as expected, after a sufficiently long time, the mean cluster size becomes time independent and reaches its saturation value $S(\kappa, \infty)$. However, some discrepancy between theory and simulation can be observed in the early times when SZT's theory is claimed to break down. We should point out that the initial condition (satisfying the excluded-volume criterion) placed particles quite close together and might have had some effect on the early evolution of the system. Although the results of SZT are not exact, they represent an asymptotic solution of the GS equation, and they seem to be very good when the system enters in the scaling regime. To analyze this point, we assume the scaling form for the size distribution suggested by Meakin and Ernst [Eq. (3)] and plot in Fig. 3 $n_s(\kappa, t) S^2(\kappa, t)$ versus $u = s/S(\kappa, t)$ for different values of $T = t\kappa^x$. In our case $\lambda = 0$, so that $z = 1$ and $x = y$. We observe that the shape of the

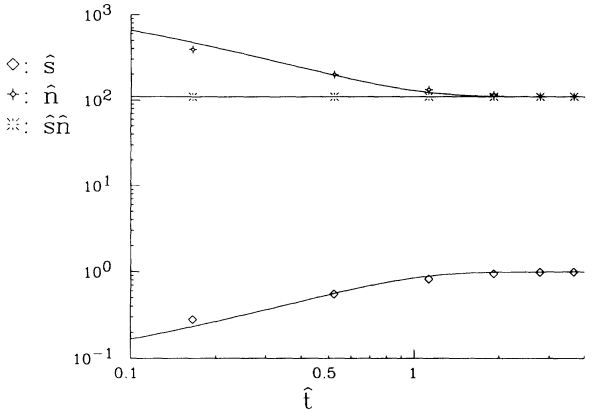

FIG. 2. Same as Fig. 1 for $\alpha = 1$.

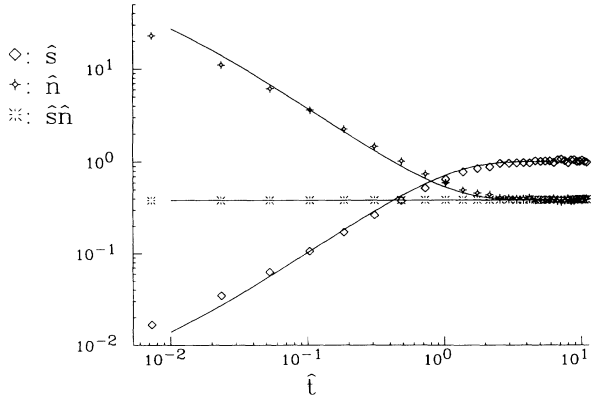

FIG. 1. Log-log plot of the reduced variables $\hat{n}$, $\hat{s}$, and $\hat{s}\hat{n}$ vs $\hat{t}$, for $\alpha = -\frac{1}{2}$ and $\kappa = 5 \times 10^{-4}$ in the three-dimensional case. Observe the good agreement between simulation data and the curves coming from solving Eq. (11).

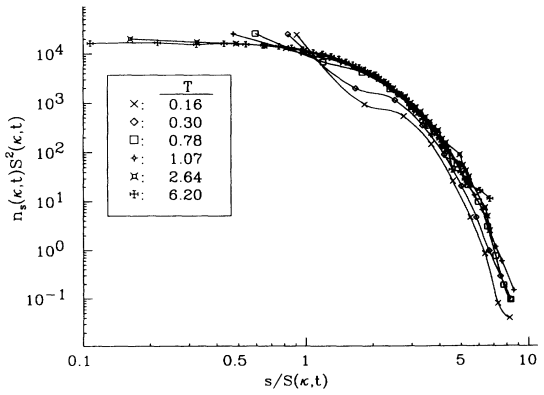

FIG. 3. Scaling of the time dependence of the cluster-size distribution. We plot in a logarithmic scale of $n_s(\kappa, t) S^2(\kappa, t)$, vs $s/S(\kappa, t)$ for $\kappa = 5 \times 10^{-4}$ and $\alpha = 1$ for different values of $T = t\kappa^x$ for the three-dimensional case.

distribution function is different at the early stages of the system evolution (i.e., for small values of T). This is because the cluster-size distribution evolves in time from the aggregation-dominated regime to the equilibrium regime, as argued by Meakin and Ernst. Presumably this causes the small differences observed between the numerical data and SZT's theoretical curve in Figs. 1 and 2.

B. Steady-state limit

To test the scaling relation, Eq. (12), we have computed the cluster-size distribution $n_s(\kappa,\infty)$, the mean size $S(\kappa,\infty)$, and the averaged number of clusters $n(\kappa,\infty)$ every 2×10^4 time steps once the system reaches equilibrium. We have averaged these quantities over 2000 measurements. To determine the exponent y we have plotted on a logarithmic scale the mean cluster size $S(\kappa,\infty)$ versus the breakup constant κ. For $S(\kappa,\infty)$ we have considered the usual definition M_2/M_1, which is denoted by $S(\kappa,\infty)_2$ in the figures. We have also considered M_1/M_0, which should be proportional to the previous measure if scaling holds, and labeled it as $S(\kappa,\infty)_1$. We have considered eight different values for κ ranging from 10^{-6} to 10^{-3}. We have also plotted on a logarithmic scale the mean number of clusters $n(\kappa,\infty)\sim S(\kappa,\infty)^{-1}$ versus κ. In Figs. 4 and 5 we compare the results obtained in two and three dimensions with the theoretical predictions, considering $\alpha=1$ in both cases. The expected theoretical value in this case is $y=\frac{1}{3}$. For the two-dimensional simulation we obtain $y=0.31\pm0.01$ from the best least-squares fit to the data. For the three-dimensional case we get $y=0.31\pm0.01$. We also include in the figures dashed lines of slope $\pm\frac{1}{3}$ coming from the mean-field theoretical result. In Fig. 6 we show the results for a three-dimensional study with $\alpha=-\frac{1}{2}$. The

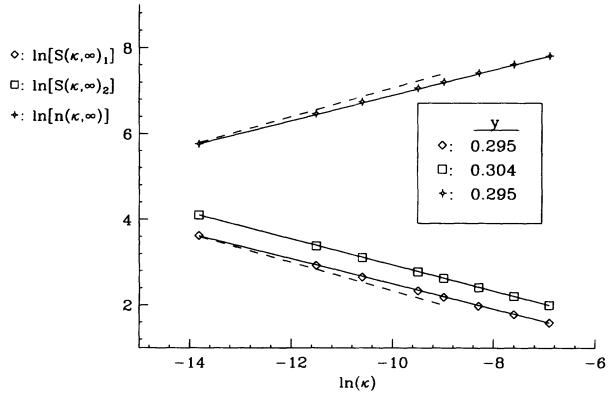

FIG. 4. Logarithmic plot of the mean cluster size $S(\kappa,\infty)$ vs the breakup constant κ, and the averaged number of clusters $n(\kappa,\infty)$ vs κ for eight different values of κ and $\alpha=1$ for the two-dimensional case. To compute the mean cluster size we use Eq. (6), which in the figure stands for $S(\kappa,\infty)_2$ and $S(\kappa,\infty)_1=M_1/n(\kappa,\infty)$. The expected theoretical value is $y=\frac{1}{3}$. Straight dashed lines of slope $\pm\frac{1}{3}$ are included to guide the eye.

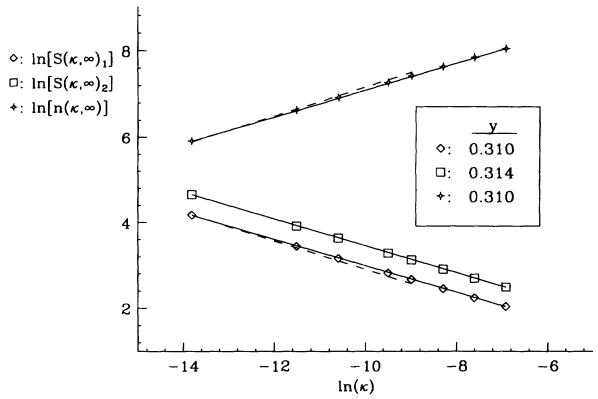

FIG. 5. Same as Fig. 4 for the three-dimensional case.

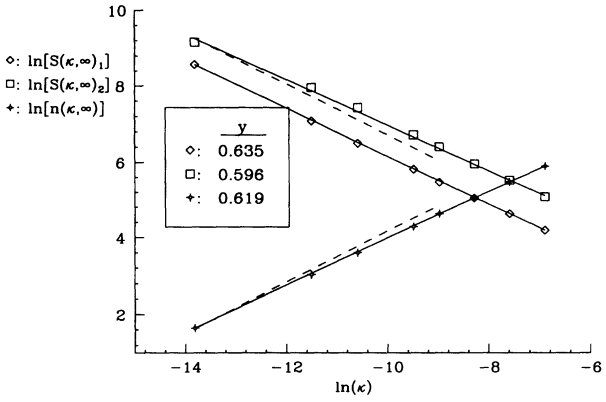

FIG. 6. Same as Fig. 5 for $\alpha=-\frac{1}{2}$. The expected theoretical value is now $y=\frac{2}{3}$.

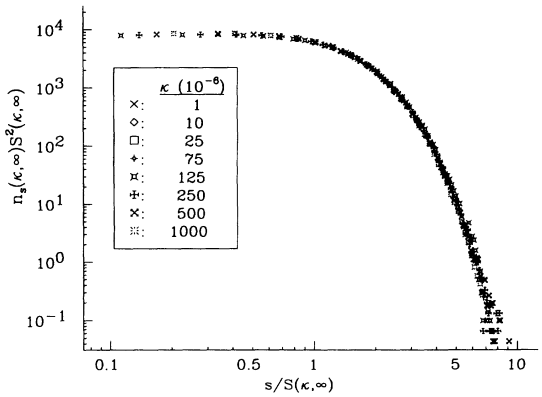

FIG. 7. Plot in a logarithmic scale of $n_s(\kappa,\infty)S^2(\kappa,\infty)$ vs $s/S(\kappa,\infty)$ for different values of κ and $\alpha=1$ in two-dimensions. Note that all data collapse reasonably well in a single master curve, supporting the scaling relation for the cluster-size distribution at the steady-state limit.

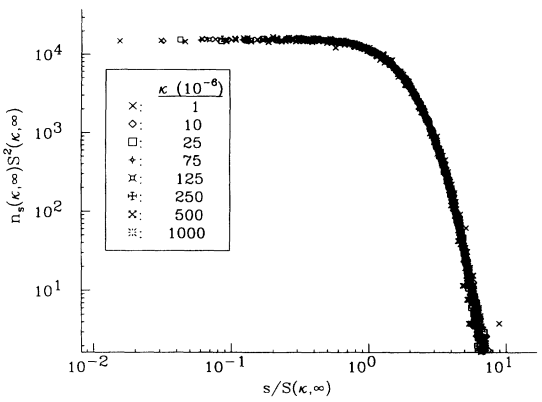

FIG. 8. Same as Fig. 7 for the three-dimensional case.

theoretical value now is $y = \frac{2}{3}$. In this case our best result corresponds to $y = 0.64 \pm 0.02$. To stress the relationship (4) at equilibrium ($t \to \infty$) we have plotted on a logarithmic scale $n_s(\kappa,\infty)S^2(\kappa,\infty)$ versus $s/S(\kappa,\infty)$. Figures 7 and 8 show these results for the two- and three-dimensional cases, respectively, for $\alpha = 1$. We observe that all of the data collapse reasonably well onto a single master curve, supporting the scaling relation assumed by Family, Meakin, and Deutch [17].

V. CONCLUSIONS

In this paper we have studied the reversible aggregation process in a single-particle off-lattice coalescence model. We have paid attention to both dynamical and steady-state scaling behaviors. We find that the results of the numerical simulation support the theoretically predicted value of the exponent describing the dependence of equilibrium size on κ. The slight discrepancies found between the mean-field results and our numerical simulations might be due to the fact that the scaling function $n_s(\kappa,t)$ evolves from the aggregation-dominated regime to equilibrium. However, this evolution is confined to small κ/s, as Elminyawi, Gangopadhyay, and Sorensen [22] demonstrated and it may have a small effect on the evaluation of the integrals that give the parameters A and B in the theoretical study of SZT. The breakdown of scaling at the early stage of the system evolution could also mean that the system might still not have entered a scaling regime and that the memory of the initial configurations has not yet been washed out. We have also computed the time evolution of the mean size and found that the numerical results and the theoretical predictions are in good agreement. Again some small differences are observed at the early stage of the evolution process in the aggregation-dominated regime. The value for the exponent y computed in this off-lattice model agrees well with mean-field results within the errors of our numerical data. Finally, we have verified the scaling assumption for the size distribution at the steady-state limit for different values of α and κ.

ACKNOWLEDGMENTS

A.C. thanks Professor C. Sorensen for many useful discussions and a critical reading of the manuscript. Financial support from the Dirección General de Investigación Científica y Técnica (Grant No. PB-89-0424, Spain) is also acknowledged.

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