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Diffusion-limited aggregates grown on nonuniform substrates

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HIGHLIGHTS

- Patterns of DLA grown on nonuniform substrates are studied.
- Incipient percolation clusters of k-mers are used as nonuniform substrates.
- At the p_c, the DLA are asymmetrical and the branches are relatively few.
- The fractal dimension of the aggregates increases as p increases.
- This behavior is discussed in the framework of existing theoretical approaches.

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ABSTRACT

In the present paper, patterns of diffusion-limited aggregation (DLA) grown on nonuniform substrates are investigated by means of Monte Carlo simulations. We consider a nonuniform substrate as the largest percolation cluster of dropped particles with different structures and forms that occupy more than a single site on the lattice. The aggregates are grown on such clusters, in the range the concentration, p, from the percolation threshold, p_c up to the jamming coverage, p_j. At the percolation threshold, the aggregates are asymmetrical and the branches are relatively few. However, for larger values of p, the patterns change gradually to a pure DLA. Tiny qualitative differences in this behavior are observed for different k sizes. Correspondingly, the fractal dimension of the aggregates increases as p raises in the same range $p_c \leq p \leq p_j$. This behavior is analyzed and discussed in the framework of the existing theoretical approaches.

1. Introduction

Fractal growth and aggregation phenomena have attracted considerable interest in the surface science community not only because of its importance for experimentalists but also by the basic theoretical background behind it [1–11]. In particular, it is interesting to notice that the structure of the aggregates strongly depends on the dynamics of the growth process as well as the topology of the substrate where they are grown [12]. In fact, great efforts have been made to develop theories and experiments capable of predicting the connection between cluster geometry and aggregation processes. Thus, exact analytical calculation describing the growth process has been developed only for simple cases. However, analytical expressions cannot be derived in more realistic cases and Monte Carlo simulations have proven to be an adequate and powerful tool to study the problem.

T.A. Witten Jr. and L.M. Sander [1] have built up one of the most studied models of surface grown: the diffusion-limited aggregation (DLA). The interest in this model is based on the essential role that the phenomenon plays in many

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experimental situations, such as electrodeposition, fluid–fluid displacement (viscous fingering), dielectric breakdown, chemical dissolution, just to name a few. As a consequence, an increasing interest has also been devoted to enhance our understanding of the theoretical basis of growing processes [1,5–10,13,14].

Most of the contributions dealing with DLA consider the fractal aggregate growing over a homogeneous surface. In contrast, considerable less attention has been paid to the study of the influence of heterogeneous substrates on the formation of DLA clusters [12,15]. It has been an old idea to consider the heterogeneous surface where DLA are grown to be the percolation cluster [16–18]. This line of thinking has been motivated by the fact that percolation clusters are good models for random porous media and DLAs have been connected to miscible displacement of one fluid by another in such media [19]. In fact, the growth of DLA on percolation clusters has been studied by numerical simulations in several seminal contributions [19–21]. In addition, theoretical predictions based on mean fields has also been established. In both cases, the percolating species forming the percolation cluster is allowed to occupy only a single site of the lattice. Recently, the influence of surface heterogeneities on the formation of DLA has been studied as aggregates are formed on, (a) one of the simplest disordered surfaces, the patchwise heterogeneous surface [22,23] and (b) Leath percolation substrates [24].

On the other hand, the pure site and the pure bond percolation of polyatomic species have been studied by using Monte Carlo simulations [25–27]. In both cases, the dependency of the percolation threshold with the size of the element deposited was discussed. In Ref. [26], it was established that the percolation threshold exhibits an exponentially decreasing behavior when it is plotted as a function of the size of the percolating species. However, very recently, Tarasevich et al. [27] have shown a nonmonotonic behavior of the percolation threshold with the size of linear polyatomic species. Nevertheless, the problem belongs to the random percolation universality class regardless the size of the percolating object.

The main aim of the present paper is to study the growing process of diffusion-limited aggregation on substrates formed by percolation clusters of particles which occupy more than one single site. These results will be helpful in understanding the formation of clusters on nonuniform substrates in thin-film-growth processes, such as vapor deposition, molecular-beam epitaxy, and so on.

In detail, the paper is organized as follows. In Section 2 the description of the substrate used for growing the DLA will be presented. The numerical procedure to produce a pattern of DLA is introduced in Section 3, along with the definitions of the critical exponents. The results of the numerical analysis are shown in Section 4, where the connections between the fractal dimension of the DLA and the critical parameters of the substrate are discussed in terms of well known relationships [28]. Conclusions are drawn in Section 5.

2. The nonuniform substrates

Let us consider a periodic square lattice of linear size L on which k-mers (a k-mer is an object composed of k identical particles each one occupying one site of the lattice) are deposited at random. For different k-mers, one must take care that the ratio L/k remains constant in order to prevent spurious finite size effects. The procedure is as follows. A k-tuple of nearest neighbor sites is randomly selected; if it is vacant, the k-mer is then adsorbed on those sites. Otherwise, the attempt is rejected. In any case, the procedure is iterated until N k-mers are irreversibly adsorbed and the desired concentration (given by $p = (kN)/L^2$) is reached. Notice that due to the already randomly adsorbed particles blocking the area, the limiting concentration or *jamming coverage*, p_j is less than that corresponding to the close packing ($p_j < 1$). In other words, p_j is defined as the maximum value of the concentration where there are no gaps available for deposition of particles of size k. The jamming coverage depends on k and exponentially converges to an asymptotic value as $k \to \infty$ [29,30].

The central idea of the percolation theory is based in finding the minimum concentration p for which a cluster (a group of occupied sites in such a way that each site has at least one occupied nearest neighbor site) extends from one side to the opposite one of the system. This particular value of the concentration rate is named *critical concentration* or *percolation threshold* and determines a phase transition in the system. In the random percolation model, a single site is occupied with probability p. For a precise value of p_c, the percolation threshold of sites, at least one spanning cluster connects the borders of the system (indeed, there exists a finite probability of finding n (> 1) spanning clusters [31–34]). In that case, a second order phase transition appears at p_c which is characterized by well defined critical exponents.

In Ref. [27], has been reported how the percolation threshold (linear k-mers) depends on k. For small values of k, the curve rapidly decreases for values of k less than 16 while this behavior changes for larger values. This nonmonotonic behavior of the percolation threshold as a function of the segment size can be associated to the following function:

$$p_c(k) = a_0/k^{\alpha_0} + b \log_{10} k + c \tag{1}$$

being $a_0 = 0.36 \pm 0.02$, $\alpha_0 = 0.81 \pm 0.12$, $b = 0.08 \pm 0.01 c = 0.33 \pm 0.02$.

As it was mentioned, the largest cluster in the range $p_c \leq p \leq p_j$ is isolated and used as a substrate for the growing process. Then, it is important to know that the main features of such a substrate are reflected in the behavior of critical exponents. For this purpose, the fractal dimensions d_f^c and d_w^c defined below are studied.

The fractal dimension, d_f^c, of the largest cluster in the lattice can be defined as [35]:

$$s \propto \rho^{d_f^c} \tag{2}$$

where s represents the number of elements belonging to the studied cluster deposited in a circle of radio ρ.

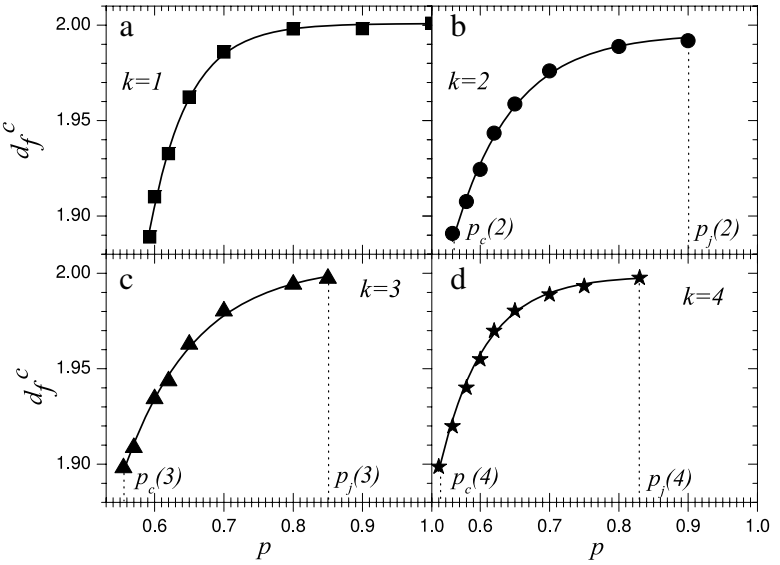

Fig. 1. The behavior of the fractal dimension, d_f^c of the largest cluster at concentration p ($p_c \leq p \leq p_j$) is shown for different values of k, as indicated. Vertical dashed lines denote the values of $p_c(k)$ and $p_j(k)$.

Fig. 1 shows the behavior of the fractal dimension of the largest cluster at concentration p ($p_c \leq p \leq p_j$) for different values of k as it is indicated. As it can be seen all the curves start from the point (p_c, 1.896) regardless the value of k. Upon increasing the concentration, d_f^c monotonically goes to 2.

Let us now suppose the motion of a particle ("an ant") which performs a Pólya random walk (unbiased, nearest neighbor random walk) on the sites of the selected cluster ("the labyrinth") at concentration p ($p_c \leq p \leq p_j$). The root mean square displacement R of the random walk is related to time t through [35]

$$R \sim t^{\nu} \tag{3}$$

where ν is a constant which depends only on the dimensionality d of the system. Here, it is important to emphasize that in Euclidean space $\nu = 1/2$ at all dimensions. A fractal dimension d_w^c is defined for the random walk by $d_w^c = 1/\nu$. For two-dimensional homogeneous surfaces $d_w^c = 2$, which is called "*normal diffusion*". However, on fractal structures R grows slower with time and d_w^c is usually larger than 2. If we consider as "the labyrinth" the incipient cluster, i.e. at concentration p_c, then $d_w^c \approx 2.73(1)$. This value is almost the same for the different k considered here (four different cases are shown in Fig. 2). As $p \to p_j$, the normal diffusion is recovered and d_w^c monotonically decreases downwards to the limit $d_w^c \approx 2$.

The determination of d_f^c and d_w^c is important for estimating the fractal dimension of a diffusion-limited aggregated growth on the above described clusters. In fact, K. Honda, H. Toyoki and M. Matsushita (HTM) [28] have discussed the influence of nonuniform space on DLA growth based on mean field theory. HTM have presented a relationship connecting the fractal dimension d_f of DLA with d_f^c and d_w^c of the cluster where the aggregate is grown on. This equation, namely

$$d_f = \frac{(d_f^c)^2 + d_w^c - 1}{d_f^c + d_w^c - 1} \tag{4}$$

has been verified to be correct only for some special cases, e.g. growth controlled by pure diffusion and ballistic motion in uniform spaces [2,12,28]. For $d_w = 2$ Eq. (4) reduces to the fractal dimension of purely diffusion-limited aggregated in the mean field approximation $d_f^{MF} = 5/3$ [1] and for $d_w = 1$ (ballistic aggregation) the above expression yields $d_f^{MF} = d = 2$ [36–38]. It becomes an additional motivation for the present paper to show the validity of Eq. (4) when the substrates are the percolation clusters discussed in the present section (which obviously are nonuniform substrates).

3. Generation of diffusion-limited aggregates

The DLA algorithm is rather simple and has been discussed in detail in the literature [1,17,39–42]. Growth begins with a seed particle in the center of the percolation cluster described above. Then, individual particles execute an unbiased random walk in such a cluster and either reach a site adjacent to the existing aggregated and stop or reach a distance far enough from the seed that the probability of a return to the aggregated is assumed to be negligible and is discarded.

Two parameters enter into the algorithm, the radius R_i at which new particles begin their random walk and the distance R_o at which they are discarded. The former becomes irrelevant as long as R_i is large enough in order to prevent the influence of the topography on the initial conditions of launching. R_i is several times greater than the maximum extent of the aggregated.

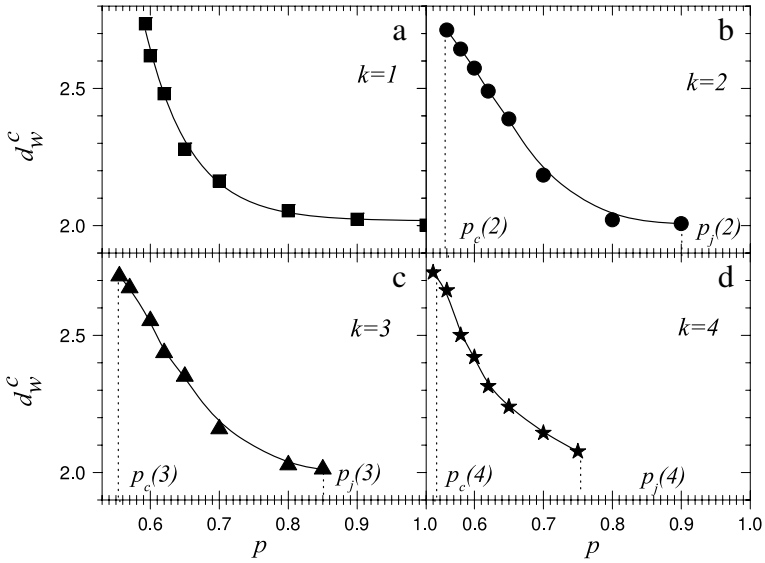

Fig. 2. The fractal dimension, d_w^c of the largest cluster at concentration p ($p_c \leq p \leq p_j$) is shown for different values of k, as indicated. Vertical dashed lines denote the limiting values: $p_c(k)$ and $p_j(k)$.

The ratio between R_o and R_i is usually a fixed number and in our simulations we have varied this quantity between 2 and 10.

Each particle launched from the circle of radius R_i performs jumps to nearest neighbor empty sites of the cluster using a random walk procedure.

The quantities reported in the present contribution have been calculated for up to 5×10^4-particles aggregated. In order to obtain accurate values of the desired quantities, averaging up to 25 different aggregates generated in the same conditions have been used.

The fractal structures are grown up to a maximum size in steps of 50 particles and the positions of the aggregated particles are recorded. At the end of such a step, the center of mass of the aggregate r_{cm}, given by

$$r_{cm}(N) = \left\langle \frac{1}{N} \sum_{i=1}^{N} r_i \right\rangle, \tag{5}$$

is determined and its radius of gyration $\xi(N)$ is calculated as

$$\xi^2(N) = \left\langle \frac{1}{N} \sum_{i=1}^{N} [r_i - r_{cm}(N)]^2 \right\rangle, \tag{6}$$

where the angular brackets denote an average over the ensemble of different aggregates. It has been shown that the "mass" N of a fractal structure is related to its radius of gyration, ξ, through

$$N \sim \xi^{d_f}, \tag{7}$$

where d_f is the fractal dimension of the DLA.

4. Results and discussions

Based on the model introduced in Sections 2 and 3, Fig. 3 shows four different patterns of DLA grown on the largest clusters formed by the deposition of tetramers with concentration (a) $p = p_c(k = 4) = 0.54$; (b) $p = 0.62$; (c) $p = 0.75$ and (d) $p = p_j(k = 4) = 0.83$ on the lattice. In the limiting case, $p = p_c(k)$, the DLA's are nonuniform and asymmetrical as compared with those grown on homogeneous substrates. In fact, they have only few branches and the structure of the DLA "follows" the cluster where they are grown on. In other words, the number of sites forbidden for diffusion (those sites of the square lattice which do not belong to the largest cluster) is close to the critical value and prevent particles from diffusion and the branches from growing. These result in the asymmetry and nonuniformity of DLA clusters that hold for values of p close to $p = p_c(k)$.

Upon increasing the probability p, the number of forbidden sites becomes less, the branch number rises and the aggregates change to relative uniform and symmetrical ones. For larger values of p (p close to $p_j(k)$) the DLA looks similar

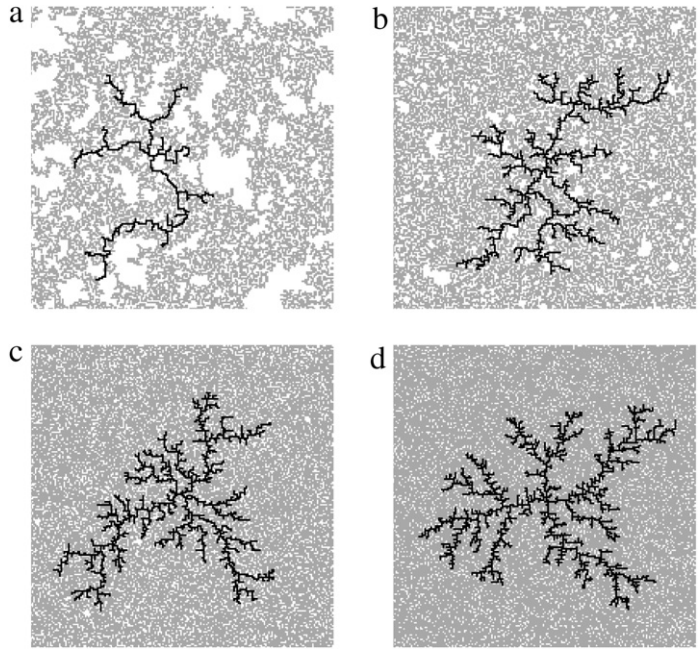

Fig. 3. Snapshots of patterns of DLA grown on percolation substrates obtained after deposition of tetramers at concentration (a) $p = p_c = 0.54$; (b) $p = 0.62$; (c) $p = 0.75$ and (d) $p = 0.83$. Black, and gray points correspond to particles belonging to the DLA and the percolating cluster, respectively.

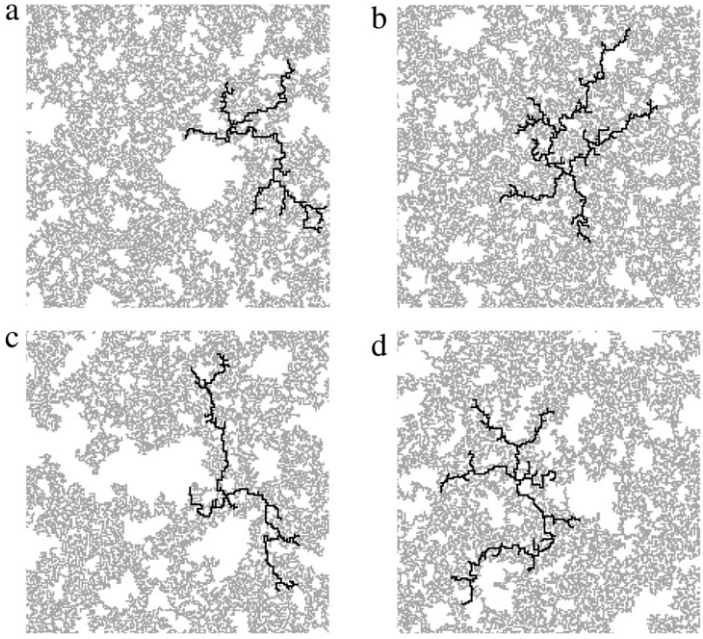

Fig. 4. Patterns of DLA grown on percolation substrates at critical coverage $p = p_c$ for clusters built with (a) $k = 1$; (b) $k = 2$; (c) $k = 3$ and (d) $k = 4$. Symbols are as in Fig. 3.

to one grown on a homogeneous lattice (in the special case when monomers are deposit on the lattice, the pattern is just as that of pure DLA).

The main difference between the DLA patterns grown on clusters formed with different k-mers is presented for concentration close to $p_c(k)$. In Fig. 4 four different aggregates are shown for (a) $k = 1$; (b) $k = 2$; (c) $k = 3$ and (d) $k = 4$ at the respective values of p_c. Since the simple observation of these snapshots (and of others not shown here) it becomes clear that only tiny differences can be found. In particular, (a) the patterns are more compact and uniform, (b) the branch number rises and its structure is more sparse and (c) the symmetry of the DLA is more similar to the pure DLA as k increases. At such a range of concentration, two competitive effects are observed in the structure of the substrate as k increases:

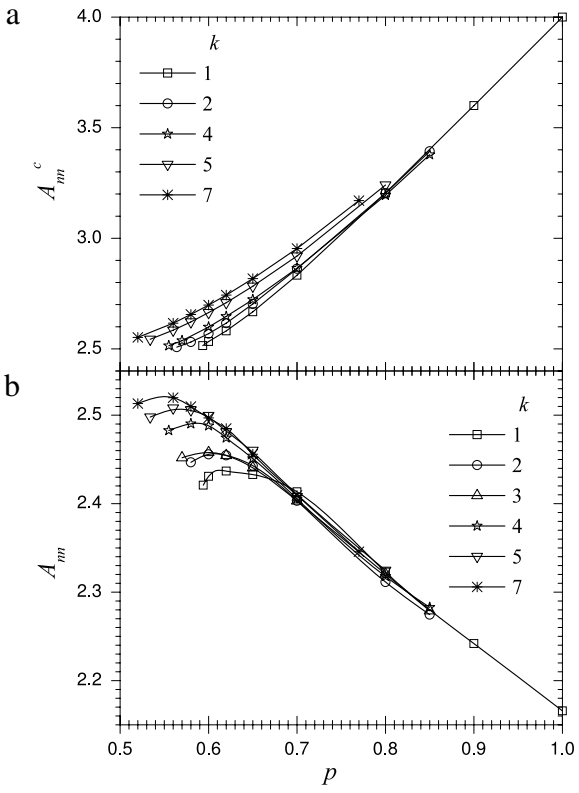

Fig. 5. The average number of nearest neighbor sites belonging to (a) the cluster, A_{nn}^c, and (b) the aggregate, A_{nn}, per site as a function of p for different values of k, as indicated.

(i) the percolation threshold (which in turn implies the number of available sites for diffusion) decreases [25,26] and (ii) the average number of nearest neighbor sites belonging to the cluster per site, A_{nn}^c, increases. In Fig. 5a, A_{nn}^c is presented as a function of p for different values of k. It is clear that only for $p \approx p_c$, A_{nn}^c varies with k. At larger values of p, A_{nn}^c tends to the same linear behavior regardless the size k. The behavior of A_{nn}^c mainly influences the structure of the DLA as it is shown by measuring the mean number of nearest neighbor sites belonging to the aggregate per site, A_{nn}, see Fig. 5b. The largest the k-mer size, the higher values of A_{nn}.

We focus now on the fractal dimension of the DLA, d_f, as a function of p for different k sizes. It is found that, in the range $p_c(k) \leq p \leq p_j(k)$, d_f increases monotonically towards a saturation value which depends on the size k. In Fig. 6, some examples are presented as full symbols. Such a behavior can be explained as follows. As it is well known in the limiting case, when $p = 1$ the fractal dimension is $d_f = 1.72$ $(k = 1)$ [43]. For different k, and p close to $p_j(k)$, the absent sites in the lattice are very few and the DLA is quite similar to the one grown on a uniform substrate. However, if k increases, p_j reaches a saturation value which in turn implies that the number of absent sites in the lattice will remains constant. As a consequence of this fact $d_f(k)$ at p_j decreases until a saturation value is reached. This is shown as squares in Fig. 7 being $d_f(\infty) \approx 1.662(3)$. Upon decreasing the probability p, the forbidden sites in the substrate increase. It in turn means the reduction of the branch number of the DLA and the decreasing of the fractal dimension, d_f. At p_c, the number of absent sites is maximum and d_f reaches a minimum value. Significantly, the fractal dimension of DLA at p_c remains almost constant regardless the value of k being a clear indication of the importance of the fractal dimension of the substrate (which is always the same $d_f^c \approx 1.896$ for the different values of k), see the circles in Fig. 7. In addition, the dashed area in Fig. 7 denotes the possible range of d_f for DLA grown on percolation clusters of particles of size k.

In order to show the reliability of the data, we analyzed the dependence of the obtained fractal dimension on the lattice size focusing on convergence. It appeared that for a given k the DLA($p = p_j$) fractal dimension does not vary much with increasing L, see Fig. 8. However, the error bars decrease with L, while the difference between the fractal dimensions of different aggregates grown on percolation clusters generated with percolating species of size k is much larger than the appropriate error. Thus, it is safe to take the values of d_f from the simulations with $L > 600$ as the asymptotic "exact" ones.

As it is mentioned in Section 2, the HTM relation has been proved in some special cases. In particular, DLA is grown on a uniform space. It is of interest to ask whether HTM is still valid for the DLA cluster growing on substrates with fractional dimension. In Fig. 6 data from Eq. (4) are plotted as full lines. From the comparison with simulation data it can be drawn that the HTM relation gives qualitatively reasonable results and a good tendency for $d_f(k)$. However, it is clear that a quantitative difference exits between numerical findings and Eq. (4) which can be estimated on average in 1.9%.

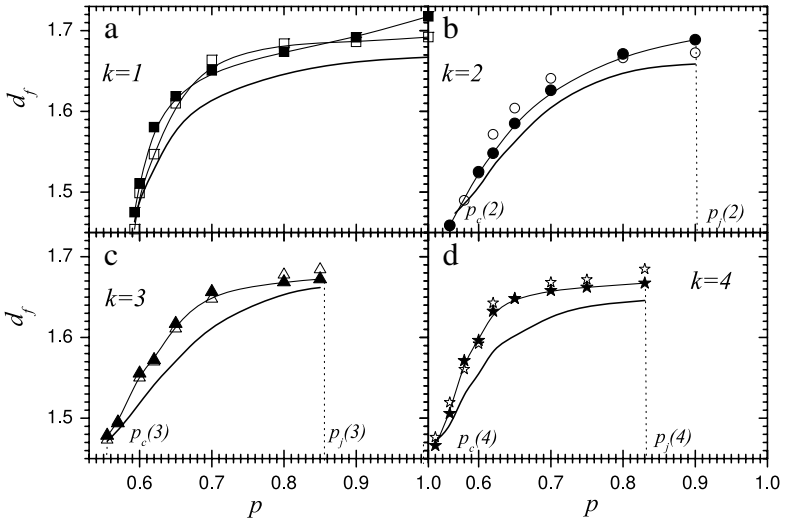

Fig. 6. The fractal dimension d_f of the DLA (full symbols) for concentration of p in the range $p_c \leq p \leq p_j$ is shown for different values of k, as indicated. Vertical dashed lines denote the limiting values: $p_c(k)$ and $p_j(k)$. Solid lines denote the HTM relation, Eq. (4) while open symbols represent the values of Eq. (8). The error in each measurement is smaller than size of the symbols.

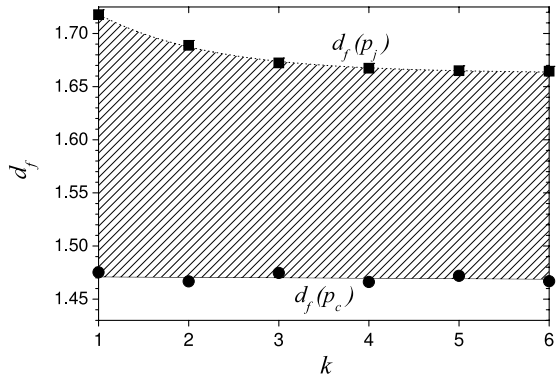

Fig. 7. The fractal dimension at p_j (squares) and at p_c (circles) as a function of k. The dashed area denotes the possible range of d_f for DLA grown on percolation clusters of particles of size k.

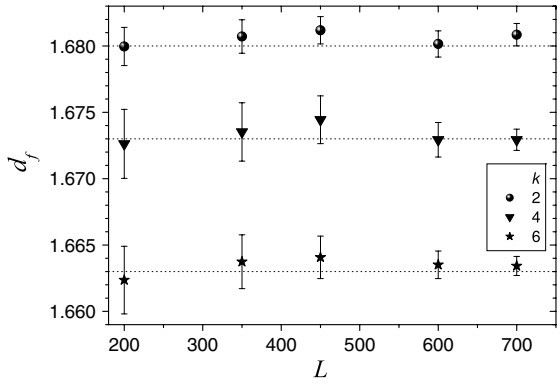

Fig. 8. Convergence analysis of the DLA($p = p_j$) fractal dimension versus the lattice size L.

In order to establish a correlation between the substrate and the DLA grown on it, we plot d_f as a function of d_f^c, see Fig. 9. Clearly, the points are very well correlated by a linear function. This tendency shows that the fractal dimension of the DLA can be expressed as

$$d_f = A + Bd_f^c \tag{8}$$

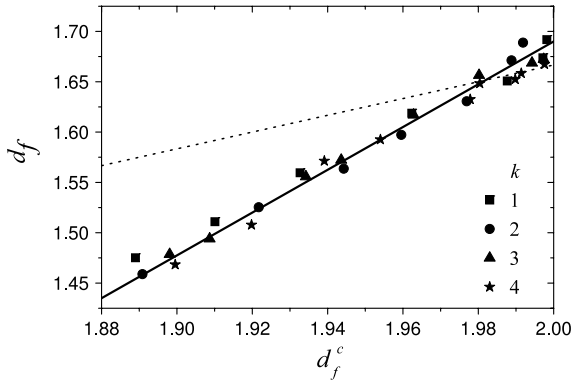

Fig. 9. The fractal dimension of the aggregates d_f as a function of the fractal dimension of the substrate d_f^c for different values of k, as indicated. The solid line represents the best fitting of the points while the dashed one is the line $d_f = 5d_f^c/6$.

where the fitting parameters are $A = -2.56(2)$ and $B = 2.125(5)$. By using the purely empirical Eq. (8) we have obtained the open symbols in Fig. 6 which differ from the numerical results by 0.55% on average. These results support the idea that the fractal dimension of the DLA depends directly on the fractal dimension of the substrate regardless the topological details introduced by the different k-mer size in the percolation cluster.

The dashed line in Fig. 9 represents the curve $5d_f^c/6$. From the figure we can see that $d_f\left(d_f^c\right)$ is different from the universal quantity $d_f\left(d_f^c\right) \approx 5d_f^c/6$, as suggested by Meakin [43,44]. A similar behavior has been reported for DLA grown on Sierpinski carpets [45,46]. Then, we can conclude that the fractal dimensionality of the DLA aggregates on the percolation cluster presented in Section 2 actually depends on more details of the lattices that are usually acknowledged, supporting the idea that the universal quantity $5d_f^c/6$ is an extrapolated hypothesis in a strict sense.

5. Conclusions

In the present paper, the diffusion-limited aggregates grown on percolation clusters formed by deposition of k-mers, for any forms, have been studied. In fact, we have considered the substrate for growing the DLA as the largest percolation cluster obtained for concentration between $p_c(k) \leq p \leq p_j(k)$. It is observed that, for $p = p_c(k)$, the DLA's are nonuniform and asymmetrical as compared with those grown on homogeneous substrates. For increasing values of p, the number of forbidden sites become less, the branch number rises and the aggregates change to relative uniform and symmetrical ones. For values of p close to $p_j(k)$ the DLA looks quite similar to one grown on a homogeneous lattice. Substrates formed with different values of k produce only tiny phenomenological differences in the DLA.

The morphology and the fractal dimension of the DLA are greatly affected by the structure and fractal dimension of the substrate where they are grown on. It is also shown that the HTM relation reproduces qualitatively the fractional dimension of the systems. This an important achievement of HTM mainly considering that it was derived from a mean field approach. However, a linear relationship between the fractal dimension of DLA and the corresponding one of the cluster gives a better quantitative agreement for the problem discussed here.

Acknowledgments

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