# Divine proportion shape preservation and the fractal nature of cluster-cluster aggregates

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We present a restricted hierarchial model of cluster-cluster aggregation which allows for an analytical calculation of the fractal dimensions in excellent agreement with those found in Nature and simulations. We argue that this agreement is a consequence of the self-preserving cluster shape common to all models and Nature. This shape determines the fractal dimension and in our model is described by *d*-dimensional generalizations of the Fibonacci series and the divine proportion. [S1063-651X(98)03212-7]

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## INTRODUCTION

When finely divided solid matter aggregates, it forms ramified clusters of apparently random and ill defined shape [1–4]. Our ability to describe the morphology of such aggregates quantitatively improved significantly some two decades ago with the introduction by Mandlebrot [5] of the fractal concept to physics, and the demonstration by Forrest and Witten [6] that these aggregates have a fractal morphology. Subsequently, there has been a resurgence of interest in the morphology of aggregates, the kinetics of their growth, and the structures that form during nonequilibrium growth in general [7–9].

Fractals are scale invariant, self-similar objects with a quantifiable, noninteger fractal dimension $D$ less than the spatial dimension $d$. The common fractal aggregates that occur in colloids [10] and aerosols [11–13] are successfully simulated on a computer using cluster-cluster aggregation models [14–16]. Despite this, fundamental questions remain with regard to how the fractal morphology occurs, and how the magnitude of the fractal dimension is determined. In this paper we present a simplified, ''stripped down'' model of diffusion limited cluster aggregation (DLCA), which explicitly shows how the fractal nature occurs and allows for an exact, analytical calculation of the fractal dimension in any spatial dimension. Remarkably, the calculated fractal dimension is in good agreement with both simulations of other models and experiment. This leads us to ask why our simple model accurately predicts these fractal dimensions; indeed, we ask what is the source of universality underlying all the various models and Nature? The key principle that arises from our model is that the shape of the aggregates is self-preserving with aggregation. In our model this shape is described by *d*-dimensional generalizations of the Fibonacci series, and determines the fractal dimension. We propose that the agreement in fractal dimension for our model and other situations is a consequence of shape preservation in all DLCA processes.

## MODEL

Our model is the on-lattice hierarchial model [12] with the following restrictions: (1) Only side-to-end collisions are allowed, e.g., on a square lattice the clusters can be circumscribed by a rectangle commensurate with the lattice; the longest edge of this rectangle is the side, the shortest is the end. (2) No part of the circumscribing rectangle of a cluster colliding with the side of the circumscribing rectangle of the second cluster can extend beyond the limits of that side. (3) The circumscribing rectangles cannot interpenetrate. This model yields a monodisperse system of clusters.

We apply this restricted, side-to-end, hierarchical aggregation scheme to the case of circular monomers on a two-dimensional, square lattice in Fig. 1. The dimensions of the circumscribing rectangles progress with aggregation in accord with the Fibonacci series

$$f_n = 1,1,2,3,5,8,13,21,34,55,\ldots . \tag{1}$$

The side-to-end aspect ratio $R$ of the aggregates is equal to $f_{n+1}/f_n$. In the limit $n \to \infty$, this yields the divine proportion (or golden section) $\phi$ [17,18]

$$R = \lim_{n\to\infty} f_{n+1}/f_n = \phi = 1.618\,03\ldots . \tag{2}$$

This simple aggregation scheme yields self-similar, fractal clusters, because with each aggregation step extra space beyond the perimeters of the combining aggregates and proportional to the aggregate size is incorporated into the new aggregate. The cluster fractal dimension for this scheme can be calculated using the facts that with each step the cluster mass increases by a factor of 2, while both the side and the

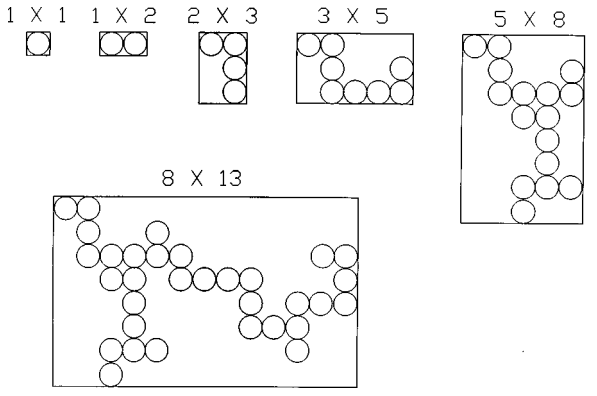


FIG. 1. A series of two-dimensional clusters created by the restricted, side-to-end, hierarchical model of DLCA. The dimensions of the circumscribing rectangles are given.

 

end of the new cluster are a factor of $R=\phi$ larger than for the two combining clusters. Thus

$$D_2=\log 2/\log\ \phi=1.44. \tag{3}$$

Here the subscript 2 refers to the spatial dimension. This value is in excellent agreement with numerous $d=2$ DLCA simulations which find, on average, $D_2=1.44\pm0.03$ [16,19,20]. The result is independent of the lattice, e.g., for a triangular lattice circumscribing parallelograms yield the same results for Eqs. (2) and (3).

To highlight the specialness of the side-to-end scheme and the importance of cluster shape consider end-to-end and side-to-side aggregations with restrictions (2) and (3) above still enforced. End-to-end aggregation yields a linear aggregate with $R\rightarrow\infty$ and a fractal dimension $D_2=1$. Side-to-side aggregation results in $R$ decreasing monotonically, if the monomers have $R>2$, until $R<2$. Then, if $a$ and $b$ are the end and side of the first $R<2$ circumscribing rectangle, $R$ will alternate between $1\leqslant b/a<2$ and $2a/b$. The geometric mean of $R$ is $\sqrt{2}$, and $D_2=2$. An alternating series of side-to-side and end-to-end aggregations creates clusters with the same aspect ratio as the clusters (or perhaps monomers) that were present at the beginning of the series. With a sufficient number of iterations, $D_2=2$ results.

In contrast, the side-to-end scheme is special in that it always yields a nontrivial fractal dimension, $1<D<d$, and a nonoscillating *shape invariance* during aggregation. If the monomers were arbitrarily shaped and could be circumscribed by rectangles of side $b$ and end $a$, then after the $n$th binary side-to-end aggregation, $R=(bf_{n+1}+af_n)/(bf_n+af_{n-1})=\phi$. This important fact demonstrates that in our simple aggregation scheme the cluster shape and fractal dimension are independent of the details of the monomers, i.e., aggregation erases the microscopic details. Moreover, the cluster shape is more than invariant; it is *self-preserving* with aggregation because aggregates or monomers of arbitrary initial shape combine to form clusters with shapes converging to $\phi$.

This aggregation scheme can be generalized to higher dimensions. The key principle is the self-preserving shape. Figure 2 shows an example of spheres on a cubic lattice in three dimensions. In $d=3$, ''side'' is the longest and ''end'' is the shortest of the three edges of the rectangular solids that circumscribe the growing aggregates. Then the progression of the magnitude of the smallest edge (the ''end'') with each step of the aggregation is 1,1,1,2,3,4,6,9,13,19,28... . This is a series similar to the Fibonacci series. Its rule is to start with three ''ones'' and then add next nearest neighbor pairs in the series to create new series members.

Reasoning by induction for an arbitrary spatial dimension we define the $d$-dimensional Fibonacci series by the rules (a) start with $d$ ''ones,'' and (b) for integer $n\geqslant d$,

$$f_{n+1,d}=f_{n,d}+f_{n-d+1,d}, \tag{4}$$

where the first subscript marks the $n$th series member, and the second subscript labels the dimension. Examples of this generalization are $d=1$, $f_{n,1}=1,2,4,8,16,...,2^{n-1}$, the geometric series; $d=2$, $f_{n,2}=1,1,2,3,5,8,13,21,34,55,...$, the Fibonacci series; $d=3$, $f_{n,3}=1,1,1,2,3,4,6,9,13,19,28,41,...$; and $d=4$, $f_{n,4}=1,1,1,1,2,3,4,5,7,10,14,19,26,36,50,...$ . Each series has a limiting value for the ratio of consecutive members

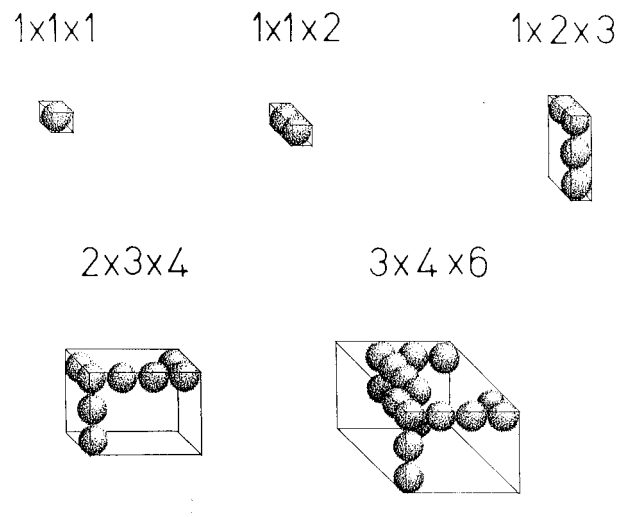


FIG. 2. A series of three-dimensional clusters created by the restricted, side-to-end, hierarchical model of DLCA. The dimensions of the circumscribing rectangular solids are given.

$$\phi_d=\lim_{n\rightarrow\infty}\frac{f_{n+1,d}}{f_{n,d}}. \tag{5}$$

This is a generalization of the divine proportion. Moreover, the self-preserving shape of the $d$-dimensional circumscribing rectangular solids implies that

$$\phi_d^d-\phi_d^{d-1}-1=0. \tag{6}$$

In $d$ dimensions, as for $d=2$, each aggregation in the restricted hierarchical model increases the number of monomers per cluster by 2, whereas the edges all increase by a factor of $\phi_d$. Thus Eq. (3) generalizes to

$$D_d=\log 2/\log\ \phi_d. \tag{7}$$

In Table I, we list $\phi_d$ values, calculated fractal dimensions, and fractal dimensions determined from simulations for Brownian, DLCA, aggregation. Agreement is quite good at all spatial dimensions, well within the error of the simulations in all instances except one. Furthermore, the calculated value in three dimensions is in excellent agreement with

TABLE I. Spatial dimension $d$, $d$-dimensional divine proportion $\phi_d$, calculated fractal dimension $D_d$(calc) from Eq. (7), to be compared to fractal dimensions from simulations of DLCA aggregation of clusters in a box $D$(box) [20], and the hierarchical model $D$(hierarchical) [19].

| $d$ | $\phi_d$ | $D_d$(calc) | $D$(box) | $d$(hierarchical) |
|---|---|---|---|---|
| 2 | 1.618 | 1.44 | $1.46\pm0.04$ | $1.42\pm0.03$ |
| 3 | 1.465 | 1.81 | $1.82\pm0.10$ | $1.78\pm0.05$ |
| 4 | 1.380 | 2.15 | $2.10\pm0.15$ | $2.04\pm0.08$ |
| 5 | 1.324 | 2.46 | $2.35\pm0.15$ | $2.30\pm0.20$ |
| 6 | 1.285 | 2.76 | $2.65\pm0.25$ | ⋯ |

naturally occurring clusters which have values near $D=1.8$ [21]. For higher spatial dimensions, there is a tendency for the calculated values to be slightly larger than, but within the error of, the simulation values. It is known, however, that $D$ from finite sized simulated clusters is smaller than that expected for infinitely large clusters. The simulation $D$ values quoted in Table I are derived from $N\rightarrow\infty$ extrapolations, which may not be sufficient.

Two remarks can now be made. First, the necessary rule for the $d$-dimensional Fibonacci series is only Eq. (4), part (b). Any series of $d$ numbers will, once part (b) is applied, yield the same asymptotic $\phi_d$. This is, again, a mathematical demonstration of the convergence to a self-preserving shape.

Second, modification of the side-to-end model to include an effective overlap between aggregating clusters results in clusters of smaller aspect ratio and greater fractal dimension. This cluster-cluster penetration is thought to describe ballistic and chemical aggregations [22], which are known to have larger $D$ values than Brownian DLCA.

## DISCUSSION

To test the predictions of our model against a realistic aggregation we have created $d=2$ DLCA aggregates by using the standard algorithm for DLCA developed by Meakin [14] which involves random aggregation of a polydisperse system of aggregates. In our simulation, $10^4$ monomers were placed at random on a $1000\times1000$ square lattice. The effective diffusion constant of the clusters was inversely proportional to their radius. Thus this simulation closely imitates real aggregation. Figure 3(a) shows the cluster radius of gyration $R_g$ versus number of monomers per aggregate. The linearity of this log-log plot is in accord with $N\propto R_g^D$, and the inverse of the slope yields the fractal dimension $D_2=1.46\pm0.03$, typical of many previous results. Figure 3(b) shows the side-to-end ratio as a function of $N$. Several clusters may have the same $N$, and for these the average aspect ratio is plotted. Two features are seen. First a broad distribution for the aspect ratio is seen with no dependency on cluster size $N$, i.e., the system has a self-preserving shape during aggregation as we have proposed. Second, the average aspect ratio is $\langle R\rangle=1.51\pm0.06$, which is too low to be described by the divine proportion, $\phi=1.618$. We remark that we have also calculated the ratio of the cluster's principle radii of gyration, and found results consistent with previous work [23,24].

The restricted hierarchical model we have presented is unlike a real aggregating system composed of polydisperse clusters coming together at random. To varying degrees similar criticism can be made for the variety of DLCA computer model systems, including aggregation of same size pairs without restrictions 1–3 above (the normal hierarchical model), aggregation of random cluster pairs and aggregation of clusters in a box allowed to randomly aggregate via various rules of aggregation, all on or off lattice. Despite these various degrees of reality, all these simulation models and now our analytically solvable model yield essentially the same fractal dimension [16,25]. This fact begs the question ''what is the common factor among all these situations that yields the same fractal dimension?'' From our simulation it appears that it is not the same aspect ratio for each model,

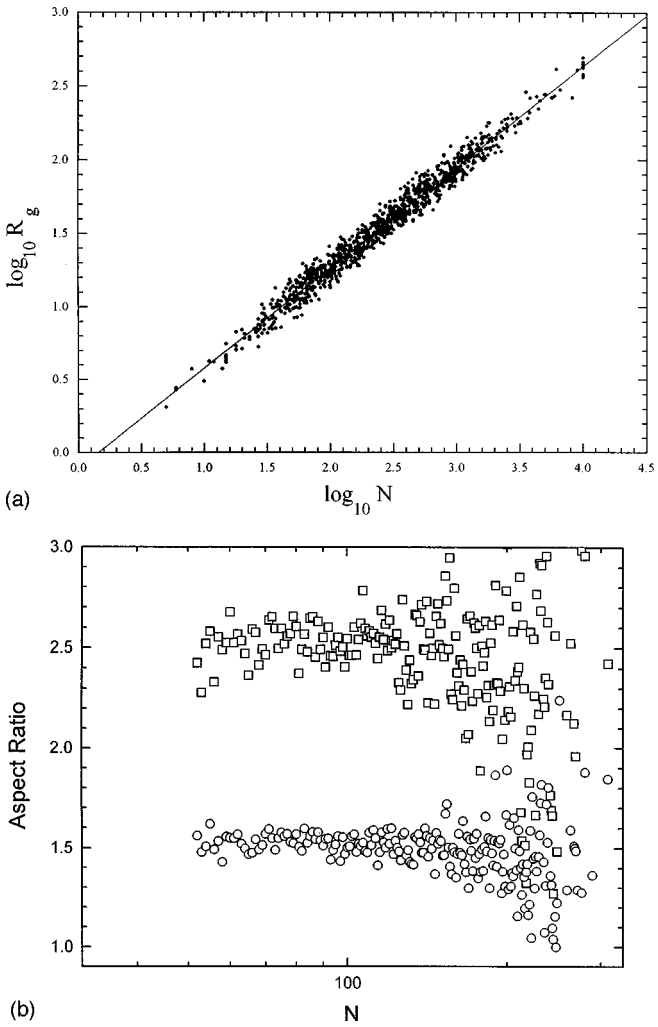


FIG. 3. (a) Cluster radius of gyration vs number of monomers per cluster for a DLCA aggregation simulation in two dimensions on a square lattice. (b) Aspect ratio of the aggregates in (a) as a function of the number of monomers per aggregate. Circles are the on-lattice length to width ratio; squares are the square root of the ratio of the squares of the on-lattice principle radii of gyration.

but it could well be the self-preserving nature of the aspect ratio with aggregation. This follows from the facts that only side-to-end aggregations in our model yield nontrivial fractals and a self-preserving shape, and our simulation and those of others show that the shape is invariant with aggregation. Thus one could argue as follows: All DLCA models have the same fractal dimension, and all models have a self-preserving shape. One model, the restricted hierarchical model, allows for a calculation of the shape and the fractal dimension. Other models do not allow for such a calculation, but since they have the same common factor the result of a calculation for the fractal dimension with one model is good for all models.

This rationalization is, admittedly, weak; but the agreement in Table I is strong; hence we scramble for an explanation. In more detail we speculate that in some manner the concept of side-to-end collisions must dominate the morphology of the resulting clusters of any DLCA model. Restricting the situation to a lattice, we can argue that an ensemble of large aggregates is the result of a random series of end-to-end, side-to-side, and side-to-end aggregations with

probabilities binomially distributed as $P_{ee}=P_{ss}=\frac{1}{4}$ and $P_{se}=\frac{1}{2}$. The equality $P_{ee}=P_{ss}$ combined with the shape replication of a consecutive pair of these, implies that side-to-end aggregations determine the morphology. This plausibility argument is edifying, but does not give an undeniable reason for the success of the model.

We end with a speculative discussion of our fractal growth model and its possible relation to other nonequilibrium growth phenomena in Nature. It is well known that many organisms follow a pattern of growth which has as its mathematical foundation the Fibonacci series and the divine proportion [17,26]. For example, the pattern of scales on pineapples and pinecones, and the inflorescence of sunflowers are intertwining spiral structures, the parastichies, the number of which is a Fibonacci number. The equiangular (or logarithmic) spiral structure of a sea shell, perhaps the most famous example of which is the shell of the Nautilus, can be based on a series of golden polygons such as the golden rectangle which has the divine proportion $\phi_2=1.618...$ as the ratio of length to width.

The results we present here imply a link between shells, pineapples, sunflowers, etc., and cluster-cluster aggregates. This link is that in all cases growth occurs by addition of material without modification of the previous structure in *a manner which creates a new structure identical in shape to the previous ones*. In his classic treatise, *On Growth and Form* [26] Thompson, who discussed the growth of a variety of shape self-preserving biological structures states that they grow ''by accretion of accumulated material'' and ''the parts, once formed, remain in being, and are thenceforth incapable of change.'' This applies to cluster growth as well. Quoting Thompson again, we may use his words intended for a shell; he writes ''...that it shall widen and lengthen in the same unvarying proportions: and this simplest of laws is that which Nature tends to follow. The shell...grows in size, *but does not change in shape*...'' This ''simplest of laws'' applies to DLCA aggregates and shells alike. What appears to be new, however, with the clusters, not found, so far as we know, elsewhere in Nature is the generalization of the $2d$ mathematics, the Fibonacci series and the divine proportion, to higher dimensions.

## CONCLUSIONS

In summary, the restricted, side-to-end hierarchical aggregation model yields clusters with a self-preserving shape described by $d$-dimensional generalizations of the Fibonacci series and the divine proportion. The cluster fractal dimensions can be calculated from the divine proportions, and the fractal nature results from incorporation during aggregation of extra volume proportional to the total cluster volume. These fractal dimensions are in good agreement with those found in Nature, and with simulations. The common factor that determines the fractal dimension for all these aggregation schemes is the self-preserving cluster shape. Finally, the simple rule that nonequilibrium growth yields structures of unvarying proportion applies to a great variety of phenomena in Nature from sea shells, to flowers, and to DLCA aggregates as well, and the underlying mathematical description for each is the same.

## ACKNOWLEDGMENTS

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