Fractal structures in 2-dimensional diffusion-limited aggregation simulations

Candidate Number: 24581
Department of Physics, University of Bath, Bath BA2 7AY, United Kingdom
Date submitted: 23 Mar 2022

Abstract

Diffusion-limited aggregation (DLA) is an important cluster formation model that has been studied extensively since its proposal in 1981. An implementation of the DLA algorithm is analysed and extended to use a non-unitary sticking probability and the presence of an attractive force between the cluster and free particle. The fractal dimension of the base DLA algorithm was found to be 1.73 which is in close agreement with the literature value of 1.71. The non-unitary attachment probability was found to follow an exponential relationship previously proposed by Bayirli. The attractive force model also followed the same linear relationship as the base algorithm but produced a much lower fractal dimension of 1.39.

1. Introduction

Fractals are self-similar or scale invariant mathematical objects. There is some disagreement on the exact definition of a fractal, but the consensus is a set which has a fractal dimension greater than the topological dimension [1]. Well-known examples of fractals are the Mandelbrot set or the Koch snowflake, which are infinitely self-similar. An infinitely divisible shape cannot exist in nature so fractals can be used as a model for certain physical processes, but not a perfect description. An example of a physical self-similar structure modelled by a fractal are Lichtenberg figures created by dielectric breakdown. These can be created as art pieces, or naturally in fork lightning.

The structures formed by diffusion-limited aggregation (DLA) processes can be treated as fractals for many purposes [2]. This scale-invariance of the structures is a useful property to exploit as properties derived from a small simulation can be scaled up to the large macroscopic scales seen in practice. The particles undergo Brownian motion and cluster together to create self-similar structures known as Brownian trees. This theory of aggregation was first proposed in 1981 by T.A. Witten Jr. and L.M. Sander [3]. Studying this model is conveniently performed using computer simulations as the algorithm is relatively simple and scalable to many dimensions [2]. This model is also very extensible, the random walk can be adjusted to simulate various other properties of materials, for example: inter-molecular forces and non-unitary likelihood of attaching for any given collision with the cluster. Variations of this model have also been used to simulate biological [4] and crystalline [5] structures.

The DLA model is of particular importance in the field of energy storage. The main factor limiting energy density in storage capacitors is the breakdown strength [6], this dielectric breakdown is modelled using DLA. High density energy storage is becoming a hot topic due to the increasing use of renewable energy and electric vehicles [7].

The model has been very well studied and its various properties analysed in the years since its introduction. This means validating an implementation of the model is simple as there are many literature sources on the expected outcomes of the simulation.

In this study the fractal dimension of the generated structures will be calculated and compared when the model is altered. The effect of increasing the number of particles in the cluster, a non-unity sticking probability, and a very basic inter-molecular force model between the 'free' particle and the cluster will be analysed.

2. Methods

The DLA model as implemented here is based on square particles confined to a grid. An initial particle is fixed at the origin of the grid, and there are three circles constructed around the cluster. The cluster circle defines the maximum radius of the cluster from the origin. The add circle is initially defined as a radius of 10, then updates to 20% bigger than cluster radius or cluster radius + 5, whichever is larger. The kill circle is initially defined as twice the radius of the add circle and on subsequent particle additions it changes to 70% larger. Particles are added at a random point on the add circle and then make random jumps in one of four directions: up, down, left or right. If the free particle has a cluster particle in either of these directions it will stick and be added to the cluster. If the particle reaches the kill circle it will be removed and replaced at the add circle. The kill circle is necessary to greatly reduce the simulation time with little impact on the cluster properties.

The process above will repeat with new particles being added to the simulation and sticking to the cluster until a predefined maximum number of particles is reached when the simulation stops. Due to the limited processing power available and the large amounts of time needed for more particles the number of particles is kept low, between 1000 and 5000.

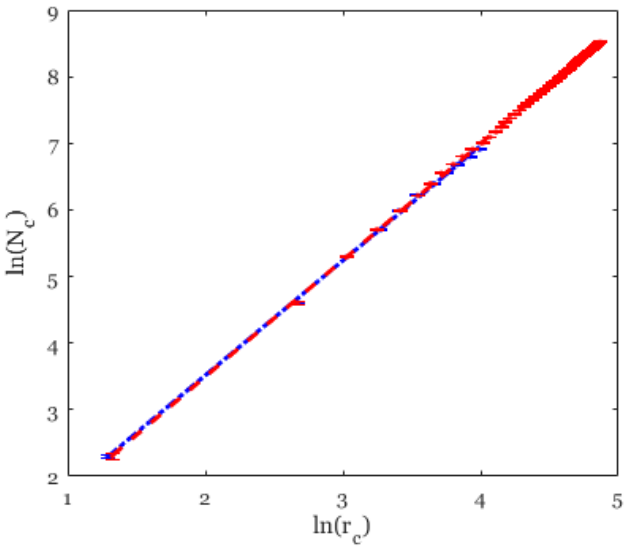

Figure 1. Log of number of particles against log of cluster radius. The blue data represents the 1000-particle data, and the red data represents 5000-particle data. The linear fits are drawn as dashed lines due to overlap. The error bars in the latter points make the data indistinguishable from the fitted curve.

The program can use a non-unitary sticking probability, which will only add the particle if it contacts the cluster and a random number generated is below the given probability. This is repeated for each side in contact with the cluster, if there are multiple. If the stick is unsuccessful the particle will continue to move, this creates the possibility of the particle attempting to move into an occupied grid point. By default, this results in the movements being rejected until a valid movement is requested and the particle continues until the stick is successful.

It is also possible for the particle to be attracted to the cluster when it comes within a certain range, simulating inter-molecular forces. This is implemented by having the particle check if it is within range of the last 10 particles to be placed. If it is out of range the particle continues Brownian motion, if it is within range it will move towards the other particle and attach.

As well as the four basic directions, the particle can be allowed to move and attach to all surrounding eight grid cells. The effect of these additional degrees of freedom on the cluster properties will also be analysed.

3. Results and Discussion

The number of particles and the cluster radius was recorded after 10 and every 100 particles added to the cluster. The following relationship

$$\frac{d\left(ln(N_c)\right)}{d\left(ln(r_c)\right)} = \frac{d_f}{1 + \beta/(\alpha r_c)^{d_f}}, \quad (1)$$

is derived assuming the number of particles N_c is proportional to the radius of the cluster r_c raised to the power of the fractal dimension d_f with a multiplicative α and additive β constant. The denominator on the right-hand side converges to one as r_c becomes large. Therefore, the constants α, β do not need to be determined and the fractal dimension can be found by plotting the log of particle number against log of cluster radius and taking the gradient to be d_f.

3.1 Calculating fractal dimension

Initially the simulation was set to only add 1000 particles to the cluster, constrain their movements to four directions, not simulate intermolecular forces and have a unitary probability of sticking. The simulation was then also run for 5000 particles.

Figure 1 shows the data for both the 1000 and 5000-particle simulations. The gradients for both graphs are very similar: d_f=1.73±0.03 for 1000 particles and d_f=1.75±0.008 for 5000 particles. The similarity between the results is to be expected as the particle runs had identical settings, just differing in the amount of data generated. This means it is likely that the 5000-particle data is closer to the true fractal dimension of this simulation. The literature value for the fractal dimension of a 2d DLA simulation is 1.71 [8], but this value is for particles unrestricted by a lattice. The restriction to a grid lattice will have minor effects on the fractal dimension. The intercepts for both plots were very close to zero with the origin well within their uncertainty ranges, validating the use of equation (1) with the large r_c approximation. The calculated dimension from these simulations agrees with the literature value of around 1.71. The discrepancy in the convergence for the 5000-particle run could be explained by the limited number of repeats (only 10) and the inherently (pseudo)random nature of the simulations as well as. The number of particles is also very low compared to what is possible on modern computers, however the available computing resources limited the number of particles and repeats that could be performed in a reasonable time.

3.2 Non-unitary sticking probability

The simulation configuration was kept identical to the first part, with 5000 particles per simulation, with the added effect of a sticking probability of less than one. The expected result of this is the cluster becomes denser as the particles can manoeuvre through the tree and attach closer to the seed particle than is possible when sticking is certain. This would cause an increase in fractal dimension as there would be a smaller cluster radius for a given number of particles. Probabilities ranging from 100% down to 1% were used which provided a wide range of cluster sizes. The relationship between fractal dimension and sticking probability has been previously modelled [9] as an exponential decay curve

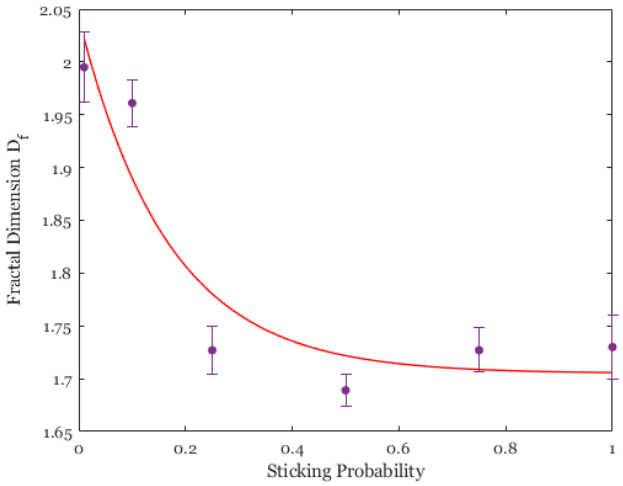

Figure 2. Fractal dimension as a function of probability of particle attaching to the cluster. An exponential decay curve is fitted as previously modelled in the literature.

$$d_f(p) = \mathrm{a}e^{-\mathrm{b}p} + d_f(1) \tag{2}$$

where a,b are simulation-dependent fitting parameters, p is the sticking probability and $d_f(1)$ is the fractal dimension for 100% sticking probability. Plotting fractal dimension against the probability and performing a nonlinear regression analysis gives the results shown in figure 2. The fitting coefficients a, b do not reveal any quantitative information about the system, but the asymptote as x->infinity gives the DLA fractal dimension which in this case is 1.71\pm 0.13. This is the exact value expected from the literature [8]; however, the uncertainty is far higher than the calculation in the previous section. This is due to the spread of the data points around the fitted curve. The plotted points are the mean from three repeats of 5000-particle runs which may not be enough for the fractal dimensions to converge, previous work has used particle numbers of orders of magnitude greater[9,10].

3.3 8-directional movement and attachment

Notably, increasing the degrees of freedom of the particle has very little effect on the fractal dimension for 100% sticking probability. It was expected that allowing the particles to attach diagonally would cause the Brownian tree cluster to increase in size for a given number of particles, but this doesn't appear to be the case. Although qualitatively, the produced structures do look slightly different due to the presence of gaps in the cluster which otherwise would not be possible. The fractal dimensions for lower sticking probabilities shown in Figure 3 increase less sharply than those in Figure 2. This could be due to the particles having double the number of directions to attach during movement than the method in Section 3.2 which offsets the lower probability somewhat. When a regression analysis fit is performed using equation (2) once again, the fractal dimension was found to be 1.72\pm 0.17 which is the correct fractal dimension but again with a large uncertainty due to the spread of the points. This is also likely to be due to the inherent randomness present in the algorithm coupled with the limited particle number.

3.4 Intermolecular attraction

Implementing an attraction between the free particle and cluster causes a massive decrease in fractal dimension for the DLA simulation. The linear relationship given by equation (1) in the limit of large r_c still appears to be valid as the intercept of the graph lies at the origin and the points follow a straight line. The results are summarized in figure 4. The fractal dimension of this model is d_f=1.39\pm 0.05. This decrease in fractal dimension is due to the particles only being attracted to the last few placed particles which will be on the limbs of the Brownian tree. This results in the limbs getting longer with very little branching occurring. This decreases the fractal dimension as there are less particles required to create a cluster of a given radius. The errors in the data points here are much larger due to the far greater range in values from the simulation. This model is far more sensitive than the regular DLA model to initial conditions as the position of a new particle placed on the starting circle has a direct effect on subsequent movements of the free particle due to the attractive force.

This model of attraction is very oversimplified, so it is unlikely to accurately represent actual processes. Allowing the free particle to be attracted to any part of the cluster would be more realistic, however, calculating distances for 1000s of particles every step causes performance issues. A way of rectifying this would be to implement a k-d tree[11] or another nearest neighbour algorithm. This would allow efficient calculation of the closest part of the cluster and could accurately simulate an attraction. The decrease in fractal dimension has been shown by more advanced analysis of attraction in previous work[12], however the fractal dimension was still higher (around 1.5) than found here.

Another possibility would be to implement a repulsion algorithm to simulate the interaction between likely charged particles (Coulomb potential) where the strength of the force is dependent on the separation of the particle and cluster [12]. This would probably require a complete rewrite of the algorithm as it currently has no implementation of momentum.

4. Conclusions

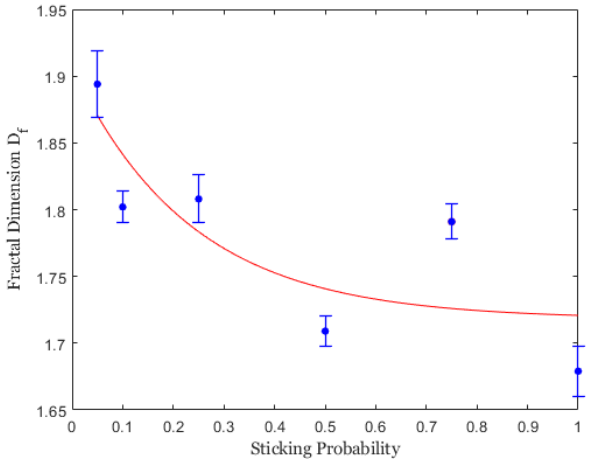

Figure 3. Fractal dimension as a function of probability of particle attaching to the cluster for particles free to move to any of the 8 surrounding grid points.

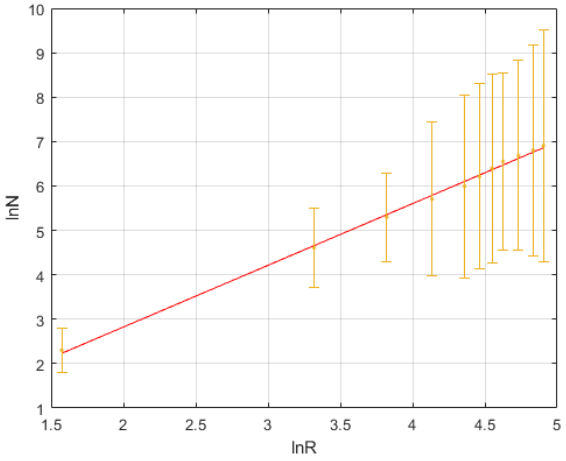

Figure 4. Logarithmic plot of number of particles against cluster radius for 1000 particles attracted to the cluster extremities. Linear regression of equation (1) form shown as red line.

It has been shown that the fractal dimension of the DLA model is around 1.7 as expected from previous work in the field. The model has become well understood since it's introduction so validating a new implementation of the model against previous work is straightforward.

The fractal dimension of the model has been confirmed to be around 1.7 for both small and medium numbers of particles. The discrepancy between the value of 1.73 found and the theoretical value of 1.71 can be explained by the particles being constrained to a grid lattice which would not be present in the physical process. The theoretical relationship shown in equation (2) was shown to hold for the model showing that the non-unitary sticking probability was implemented correctly in the model. Increasing the degrees of freedom of the free particle by allowing it to move diagonally as well as horizontally and vertically only had a statistically significant effect for low (<50%) sticking probability. This effect was not fully accounted for and would require further research, but it could just be an artifact of the limited dataset. The simple attraction model was shown to greatly decrease the fractal dimension to 1.39 which was accounted due to the particles only being attracted to the last few particles to be placed which would cause limb-lengthening of the Brownian tree.

Further development of the attraction model and either an optimization of the DLA algorithm implementation or greater computer resources would be necessary to test larger clusters with more particles. More data with more particles would allow for the random spread in the data caused by the pseudorandom motion of the particles to be greatly reduced and hence the confidence in the calculated values and the conclusions drawn from them would increase. Allowing for shapes other than squares, like circles could also be implemented which could give a more realistic depiction of atomic interactions.

Increasing the number of spatial dimensions in the algorithm from 2 to 3 would increase the complexity greatly but would allow simulation of many more physical DLA systems.

References

[1] Mandelbrot B 2004 *Fractals and Chaos* (Springer)

[2] Ball R, Nauenberg M and Witten T A 1984 Diffusion-controlled aggregation in the continuum approximation *Physical Review A* **29** 2017

[3] Witten T A and Sander I M 1981 PHYSICAL REVIEW LETTERS Diffusion-Limited Aggregation, a Kinetic Critical Phenomenon **47**

[4] Caserta F, Eldred W D, Fernandez E, Hausman R E, Stanford L R, Bulderev S v., Schwarzer S and Stanley H E 1995 Determination of fractal dimension of physiologically characterized neurons in two and three dimensions *Journal of Neuroscience Methods* **56** 133–44

[5] Gentile F, Coluccio M L, Toma A, Rondanina E, Leoncini M, de Angelis F, Das G, Dorigoni C, Candeloro P and di Fabrizio E 2012 Electroless deposition dynamics of silver nanoparticles clusters: A diffusion limited aggregation (DLA) approach *Microelectronic Engineering* **98** 359–62

[6] Yang L, Kong X, Li F, Hao H, Cheng Z, Liu H, Li J-F and Zhang S 2018 Perovskite lead-free dielectrics for energy storage applications

[7] Dunn B, Kamath H and Tarascon J M 2011 Electrical energy storage for the grid: A battery of choices *Science* **334** 928–35

[8] Halsey T C 2000 Diffusion-limited aggregation: A model for pattern formation *Physics Today* **53** 36–41

[9] Bayirli M 2009 The numerical approaches of relations to the fractal dimension D and the stick probability coefficient t in the diffusion-limited aggregation *Journal of Optoelectronics and Advanced Materials - Symposia* **1** 547–50

[10] Braga F L, Mattos O A, Amorin V S and Souza A B 2015 Diffusion limited aggregation of particles with different sizes: Fractal dimension change by anisotropic growth *Physica A* **429** 28–34

[11] Bhatia N and Vandana 2010 Survey of Nearest Neighbor Techniques *IJCSIS) International Journal of Computer Science and Information Security* **8**

[12] Jungblut S, Joswig J-O and Eychmü A 2019 Diffusion-and reaction-limited cluster aggregation revisited *Phys. Chem. Chem. Phys* **21** 5723

Appendix / Appendices