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A discrete model for particle deposition

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Simulation of deposit growth on a two-dimensional substrate was studied based on a new model that tracks individual cubic particles as they form a deposit structure. The present model is an extension of the classical ballistic deposition model. Effects of three different parameters were studied. These include an attraction parameter that is a measure of the particle to particle attractions, an interaction length within which the particles are assumed to influence and be influenced by surrounding particles, and allowed sticking positions (face-face, edge-edge and corner-corner) that favor particular growth directions. Structures with widely varying properties were obtained using this model. The three parameters were found to have considerable effect on the structure including indications of morphological phase transformations. A new property of the system (saturated roughness/deposit growth rate) was identified that can classify the different types of growth into a single type.

1. Introduction

Ballistic deposition was originally proposed by Vold [1] and Sutherland [2] as a model for colloidal aggregation. This work was later extended and analyzed to simulate the process of vapor deposition, where the initial deposit structure [3] shows somewhat similar scaling behavior [4, 5]. The simulation is usually done in the following way: particles are assumed to drop along straight lines on the deposit and attach to the surface or any other particle at the perimeter site. This apparently simple rule produces a structure that is complex and has been extensively analyzed for scaling behavior. Structurally, ballistic deposits are not fractals. Careful numerical simulation [6] as well as theoretical argument [7] have verified this. However, the surface roughness of the deposit structure shows interesting scaling behavior, first investigated by Family and Vicsek [8] who quantified the roughness in terms of the surface width, or rms variation in height.

$$W^2 = \frac{1}{l^{d-1}} \sum_i (h_i - h)^2 \qquad (1)$$

where l is the system size, d the dimensionality of the substrate, h_i the height of the substrate at the ith location and h is the mean height of the substrate. The scaling behavior is characterized by two parameters, α and β. The width, W, scales as t^β in the initial stages of growth and then saturates. The saturation value of width scales with the size of the system as l^α. The scaling behavior has been studied extensively. Theoretical explanation of the scaling behavior has been proposed based on the Kardar-Parisi-Zhang approach [9].

Much work in this area has been devoted to morphological transitions in deposition models. Variations of the original model have been suggested and explored to include additional parameters and construct a more generalized model. Additional emphasis has been given to study if changes in parameter values can initiate a change in the deposit structure. Finite Density Ballistic Aggregation [10] was proposed with a parameter p that mimics an incoming flux that can be varied to alter deposition structures. Another recent work [11] suggested using two kinds of particles and extending the interaction between particles to next nearest neighbors. Both of these models have shown the existence of morphological transitions associated with a change in value of some parameter.

In the present work we have attempted to present a realistic model that tracks individual particles as they form a deposit structure. The original motivation was plugging of a control valve in an aerospace application [12]. In that problem, it was essential to understand the role of particle size distribution, loading, and sticking probability on deposit height and morphology. The traditional approach to deposition morphology problems is to integrate the equations of motions for each particle to follow the trajectory. However, because of the computational complexity, this approach is limited to a fairly small number of trajectories. In contrast, Saunders et al. [12] constructed a lattice deposition model based upon a generalized ballistic deposition approach with additional parameters representing the

microphysics, and substantial insight into the deposit morphology was obtained with rapid computational times. Simple attachment probabilities and sticking rules represented the microphysics and were found to control the deposit morphology. In the present work we have extended the approach to study three important parameters that affect the system: an attraction parameter, an interaction length that limits the range of interaction between particles, and position of sticking (face-face, edge-edge-edge and corner-corner). The dynamic scaling behavior was studied extensively along with other characteristics of the structure like rate of change of average height and porosity. It was found that the attraction parameter has a strong influence on the structure. The interaction length was found to affect the structure strongly in the initial stages. Effects of increasing this parameter to include the influence of particles at longer distances generally dies down quickly, especially in the high attraction parameter regime. The choice of a position of sticking was found to affect the structure most, altering the way in which surface properties like roughness vary with the attraction parameter in the case of regular face-face sticking.

2. The model

Deposition and sticking rules for the present model are shown in Fig. 1. The modeling is carried out on a three dimensional lattice with the substrate consisting of a two dimensional surface. The lattice cell spacing is unity in all the three directions. Individual cubic particles are released at randomly chosen positions above the substrate. The particles follow a vertical downward path and attach to surface sites following a set of rules. There are three factors that determine these rules.

(1) An *attraction parameter* that is a measure of the particle to particle attractions, such as due to electrostatic forces. It represents the attraction between two particles on a relative scale from 0 to 1, with the former representing no interaction and the latter representing the maximum interaction.

(2) The *sticking position* where one particle can attach to another (face-face, edge-edge or corner-corner).

(3) The *number of nearest neighbors* that can have an effect on the probability of sticking of an incoming particle at a site.

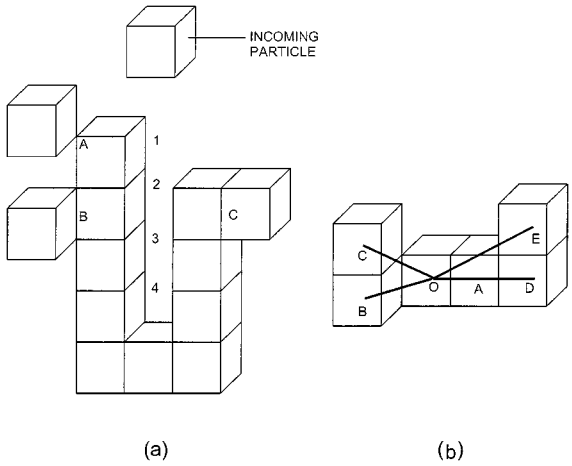

Figure 1 Summary of sticking rules: Incoming particle can occupy any one of the sites 1, 2, 3 or 4. The type of sticking can be face-face (C), edge-edge (B) or corner-corner (A) [see Fig. 1a]. The incoming particle is first tested for attachment at site 1, then 2 and so on. The presence and location of neighboring particles determine the sticking probability of the particle at a site. An interaction length is defined beyond which particles are assumed not to interact with each other [see Fig. 1b]. The number of neighboring particles is decided based on the number of particles present within this distance. For a lattice model this distance can have discrete values only. Five such lengths were investigated: 1 (length of OA in the figure), $\sqrt{2}$ (OB), $\sqrt{3}$ (OC), 2 (OD) and $\sqrt{5}$ (OE).

The different positions of sticking have been considered here to study the effect of the growth directions on the structure of the deposit. Face-face sticking involves growth in four perpendicular directions on the substrate along with vertical growth. Addition of edge-edge and corner-corner sticking makes possible growth in additional directions on the substrate plane, including some out of the plane, at 45° from the vertical. In evaluating the effect of the *number of nearest neighbors* we have considered the effect of an *interaction length* beyond which it is assumed that the mutual attractive effect of one particle on another vanishes. So, when calculating the attractive effect of the neighboring particles on an incoming particle at a particular site, we only consider the effect of those particles that are within a pre-defined interaction length from that site (measured in terms of center-to-center distance). Since we are considering a lattice model with a unit lattice parameter, where cubic particles can occupy specified positions only, this interaction length can only take discrete values (1, $\sqrt{2}$, $\sqrt{3}$, 2, $\sqrt{5}$ and so on). Additionally, assuming a Coulomb type interaction between the particles, the actual attraction between two particles is proportional to the inverse square of their center to center distance. We define an attraction parameter p for a problem, which represents the attraction in the limiting case of a face-face contact between the particles. For any distance larger than this, the attractive effect is decreased based on the inverse square of the center to center distance.

$$\frac{P_d}{p} = \frac{1}{d^2}, \quad \text{for } d \geq 1 \tag{2}$$

Also, the net effect is assumed to be additive, i.e., the final sticking probability of a particle to a site is calculated based on contributions from all particles that are within a pre-defined interaction length for the problem.

An incoming particle gets attached to a surface site based on the net sticking probability of that site. If the particle falls on a column that is higher than its neighbors, (frontal collision), it gets attached to the top of the column. On the other hand, if the particle is on a column that has higher columns in its neighborhood, then it gets attached to any one of the available positions based on their net sticking probabilities. A net sticking probability of 1 or more for a site indicates a sure attachment of an incoming particle to that site. For all other cases (between 0 and 1), attachment is achieved when the net sticking probability is greater than a random number generated from an uniform distribution between 0 and 1. If the height of the column on which the particle has been released is h_o and there are higher columns in its neighborhood, the highest of which is h',

the particle will be tested for attachment at all possible sites between h' and h_o.

The following points should be noted: (1) A valid site between h' and h_o is determined based on the location of the neighboring particles. Thus, in the case of face-face sticking, there has to be a particle that shares a cubic face with a site, for the site to be a valid one. When edge-edge sticking is also allowed, there has to be one shared edge or face between a possible site and its neighboring particles for it to be a valid site. The same rule is extended to corner sharing when corner-corner sticking is allowed. (2) A face-edge, edge-corner or face-corner sticking is not permissible in the present model since it does not allow off-lattice particle attachment and (3) an attraction parameter equal to 1 and interaction length of unit lattice distance (i.e. face-face sticking is only allowed) represents the classic ballistic deposition model.

The physical motivations for the model are manifold. We are looking for a more general model than the ones usually suggested for simulating vapor deposition. The attraction parameter, as discussed earlier, is a physically realizable quantity and can be related to system parameters like temperature, charge or even the velocity of the incident particles. The study of the interaction length was motivated by the fact that an attractive effect between the deposit and an incoming particle can be extended beyond the immediate neighborhood of the particle in many physically possible cases. Such an effect can also be related to a local minimization in the energy and hence a sort of local equilibrium (particles are more likely to stick to a site surrounded by more particles). The testing of particles at individual sites downward achieves two things. (1) It is representative of a force that drags the particles in a particular direction (such as gravity or carrier gas velocity) and (2) it minimizes particle penetration into very narrow grooves on the surfaces. To see this, consider a vertical groove of unit side-length bounded on all sides by filled columns. In reality, a particle entering the top is likely to attach to the side walls before penetrating to the groove bottom. Testing the sticking probability on the column sides downward simulates this.

The simulations were done on an 80×80 substrate with periodic boundary conditions. The number of particles used in the simulation was between 40 and 60 million. Interaction length from 1 to $\sqrt{5}$ was used in the simulation for face-face sticking. For each cut-off distance the attraction parameter was varied from 0 to 1. One series of simulations with attraction parameter varying from 0 to 1 and an interaction length of $\sqrt{2}$ was done for the case where edge-edge sticking is allowed along with face-face sticking. A similar set of simulations were done for a case where corner-corner sticking is also permitted, the interaction length used in that case being $\sqrt{3}$. A summary is given in Table I.

TABLE I Summary of the simulation conditions

Type of sticking	Attraction parameter (p)	Interaction length (d)
Face-face	0 to 1	1, $\sqrt{2}$, $\sqrt{3}$, 2, $\sqrt{5}$
Face-face and edge-edge	0 to 1	$\sqrt{2}$
Face-face, edge-edge and corner-corner	0 to 1	$\sqrt{3}$

3. Results

3.1. Average height of the deposit

The average height, h, of the deposit was found to vary linearly with time (time has been measured here in arbitrary units). We assumed a constant inflow rate of particles; hence time is proportional to the number of particles. The rate of growth, dh/dt, was found to depend strongly on the attraction parameter, for the lower values of p. A typical variation of average height with time (for an interaction length of $\sqrt{2}$ and face-face sticking) is shown in Fig. 2. Fig. 3 shows the variation of average height growth rate with attraction parameter for various interaction lengths. The change in the growth rate with p seems to follow a power law behavior. Except for the case where the interaction length, $d = 1$, all the others reach a saturation growth rate beyond a certain value of attraction parameter. The effect of sticking position is shown in Fig. 4. Clearly, there is a sudden change in the behavior of the system when edges and corners are included as possible sites for sticking. We will find that this behavior is a general one and is reflected in the behavior of other morphological characteristics also.

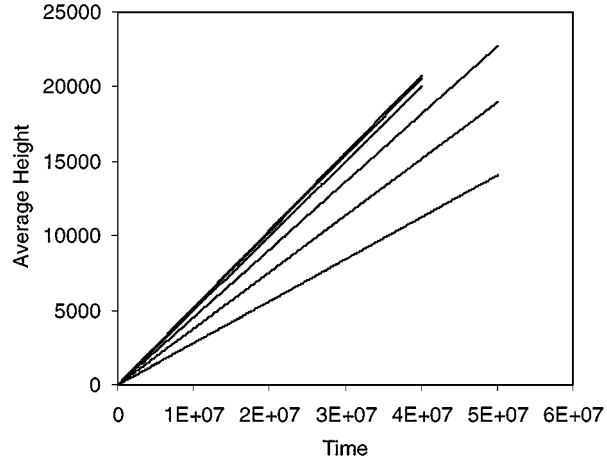

Figure 2 Change in average height with time (for face-face sticking and $d = \sqrt{2}$).

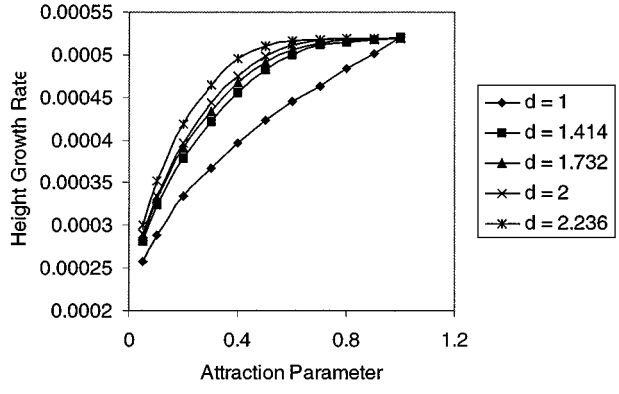

Figure 3 Height growth rate vs. attraction parameter for different values of d. The initial change follows a power law behavior and then saturates (except for $d = 1$).

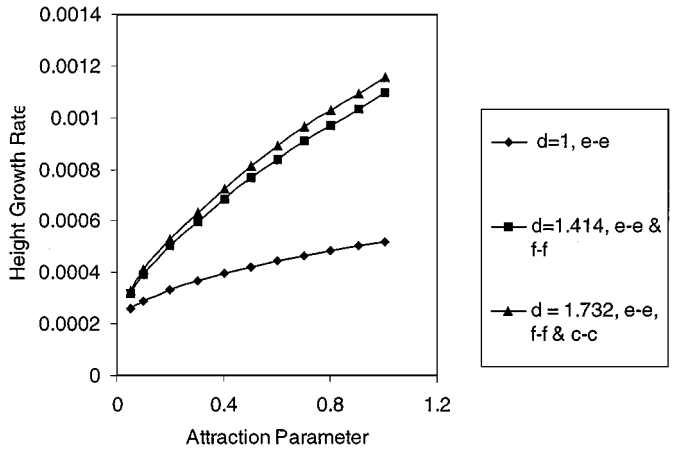

Figure 4 Average height growth rate vs. attraction parameter for different positions of sticking.

3.2. Porosity of the deposit

Typical porosity change with time is shown in Fig. 5 (for an interaction length of $\sqrt{2}$ and face-face sticking). Porosity increases rapidly and then becomes constant. The saturation porosity is found to depend on the attraction parameter as shown in Fig. 6 (for an interaction length of $\sqrt{2}$ and face-face sticking). The behavior is similar to the change in height shown in Fig. 3. In both the cases, a change in the behavior of the system is noted as the attraction parameter, p is varied. For higher values of p, the behavior remains unchanged. As p decreases, both the saturated porosity and the rate of growth of average height change. The range of p over which this happens depends on the value of the interaction length, d. As d increases, the saturation is reached at lower values of p, for both the average growth rate and the saturation porosity. Fig. 7 shows the effect of changing sticking position. A sudden change similar to one observed in the case of average growth rate is observed here, indicating that the change in sticking position also has an important effect on the morphology of the system.

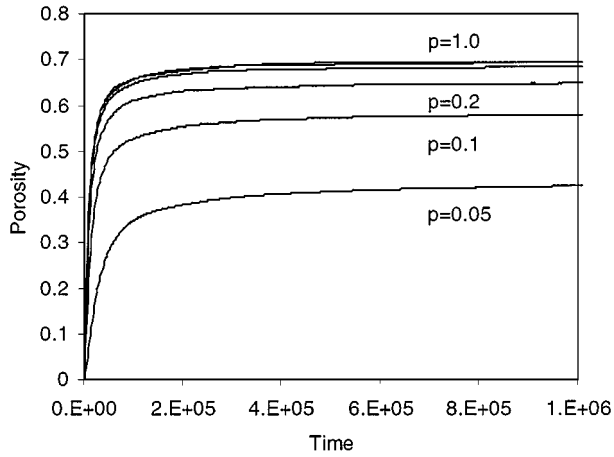

Figure 5 Change in porosity of the deposit with time (for face-face sticking and $d = \sqrt{2}$).

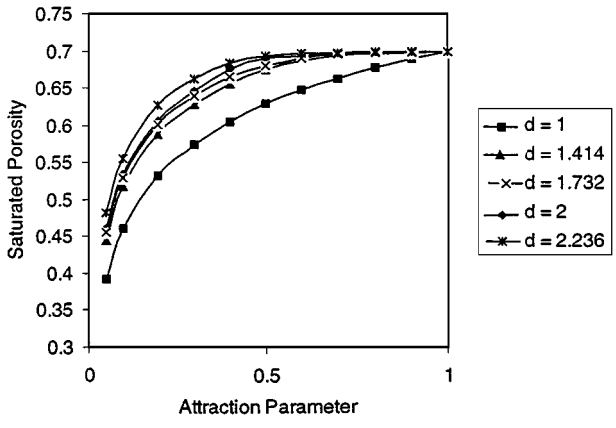

Figure 6 Saturated porosity vs. attraction parameter for different values of d.

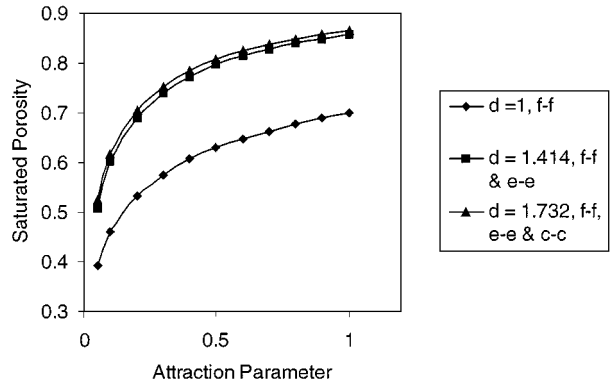

Figure 7 Saturated porosity vs. attraction parameter for different sticking positions.

3.3. Surface roughness

The roughness of the surface is characterized in the literature using two parameters, α and β, which are calculated from the width of the deposit according to Equation 1. β is the dynamical parameter that measures the initial growth rate of the width with time, given by $w \propto t^{\beta}$. α is a scaling parameter that relates width to the length scale in which it is measured. For a non-fractal structure we should not expect any variation in width as length scale is changed. In the present case we studied only the dynamical parameter since it was computationally easier to obtain.

Changes in width with time for three different sticking positions are shown in Figs 8–10. The change in behavior when edge-edge and corner-corner sticking is

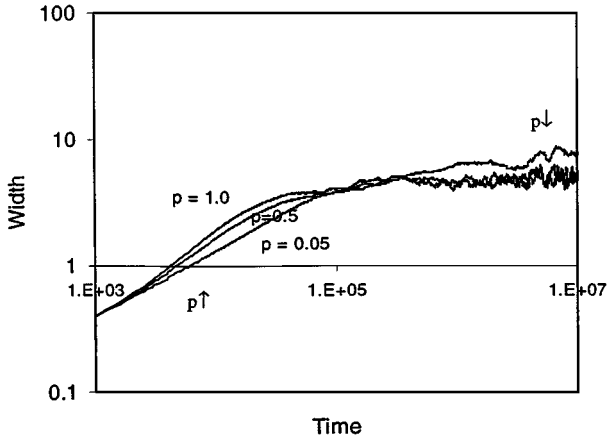

Figure 8 Change in width (roughness) with time for three values of attraction parameter (face-face sticking and $d = 1$).

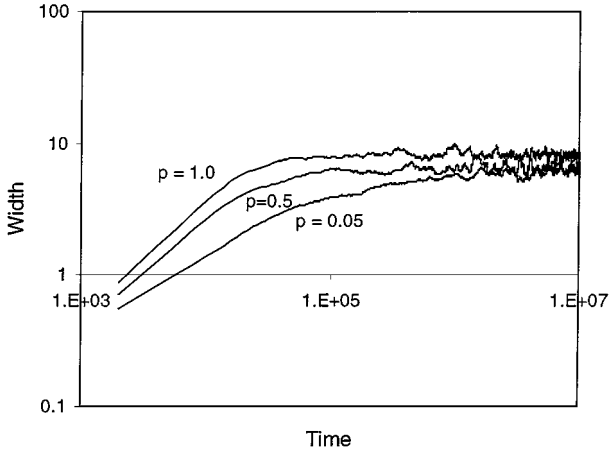

Figure 9 Change in width (roughness) with time for three values of attraction parameter (face-face and edge-edge sticking, $d = \sqrt{2}$).

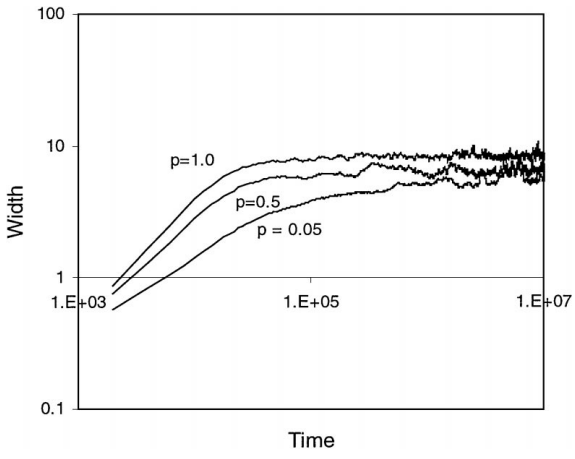

Figure 10 Change in width (roughness) with time for three values of attraction parameter (face-face, edge-edge and corner-corner sticking, $d = \sqrt{3}$).

allowed should be noted. A common feature in all the three cases is that for a fixed type of sticking position, during the initial growth period, the width or roughness increases with attraction parameter. At longer times in Fig. 8, with only face-face sticking the behavior changes: the width decreases with attraction parameter.

Initially, the increase in width with time follows a power law behavior. For the model with face-face sticking and interaction length, $d = 1$ we found that β depends on attraction parameter (p) strongly for low values of p. However, it saturates quickly when p increases as Fig. 11 shows. The saturation value is around 0.22 which matches closely with the reported values of β [10, 13, 14] for the classical ballistic deposition model. For higher d, β usually has a lower value for the same p. The saturation of β is also reached for at lower values of p. The situation is reversed when edge-edge or corner-corner sticking is allowed along with face-face type of sticking. As Figs 9 and 10 show, the β values seem to increase as the attraction parameter increases. Fig. 12 gives a comparison of the ways in which width changes with time when $p = 1$ for the three types of sticking positions. The models that allow edge-edge and corner-corner type of sticking clearly have $\beta > 0.22$. They also saturate earlier than the model that allows face-face sticking only for the same values of p. A possible explanation is that the addition of an edge-edge or corner-corner type of sticking causes the correlation distance (i.e. the "information" the height of one column passes on to a nearby column) to increase at a faster rate. For example an edge sticking on the vertical edges or any corner sticking helps in extending the interface in both x and y directions as compared to a face sticking that extends the interface in either x or y direction. So, the effect of a locally high column spreads much faster and hence the early saturation of width.

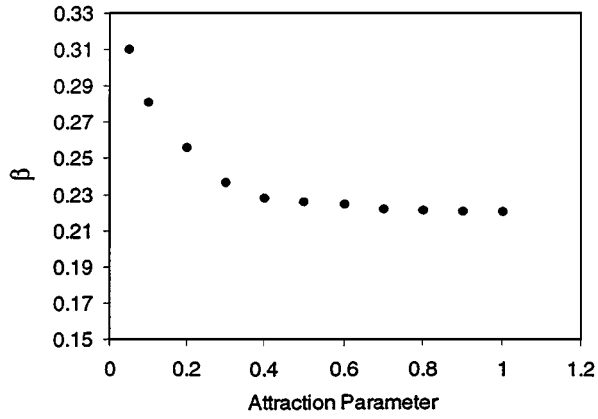

Figure 11 Change in β with attraction parameter (face-face sticking, $d = 1$).

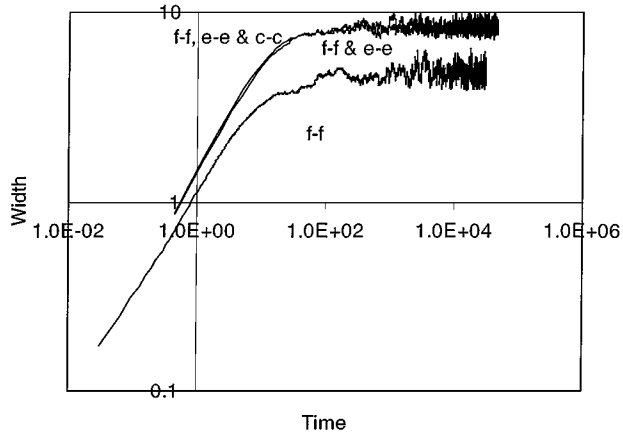

Figure 12 Change in width (roughness) with time for three different sticking positions ($p = 1$).

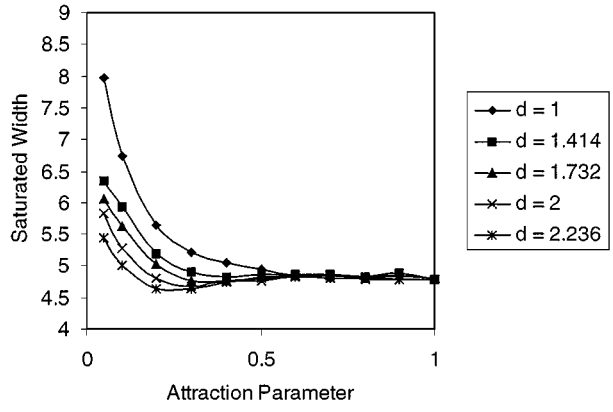

Figure 13 Saturated width vs. attraction parameter for different values of d.

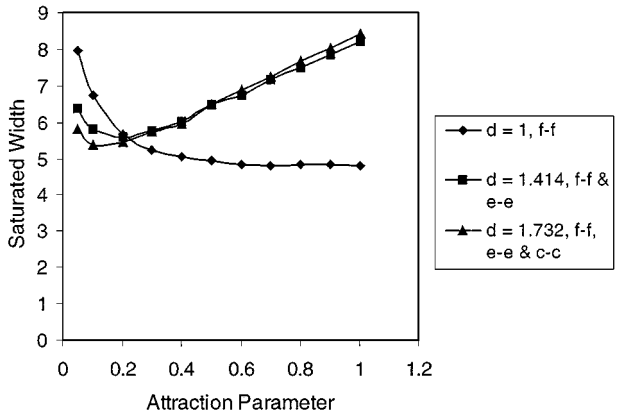

Figure 14 Saturated width vs. attraction parameter for different sticking positions.

Saturated width value (W_{sat}) gives a measure of the final roughness of the surface. Fig. 13 shows the variation of saturated width with p for different values of d and the face-face sticking only model. It seems that the attraction parameter has little effect on the structure beyond an upper critical level. This may be due to the increase in number of nearer neighbors, which causes the effective sticking probability of a site to be more than 1 in most of the cases. This is also reflected in the observation that as the interaction length increases, the saturation of surface width value occurs at lower p. The situation is reversed when edge-edge and corner-corner type of sticking is allowed. In these cases, saturated width values increase with p as shown in Fig. 14 after a small initial range of values of p where it decreases. The initial decrease may be due to the fact that for very low values of p the chances of edge-edge or corner-corner type of sticking is less (attraction parameter is reduced to $p/2$ and $p/3$ respectively for edge-edge and corner-corner type of sticking). Hence, for low values of p they are expected to have more of face-face type of sticking than any other type and hence the decrease similar to that observed for face-face sticking only type of model. For higher values of p the relative proportion of edge-edge and corner-corner type of sticking increases substantially and that causes the increase in the saturated width value.

4. Discussion

The effects of three parameters, p, d and position of sticking were studied in this work. The properties of the deposit that were studied include height, porosity and width. The change in structural properties due to changes in the system parameters indicates that morphological transitions are taking place. All the three parameters seem to effect the morphological changes.

A change in the attraction parameter with other factors like position of sticking and d remaining constant affects the structure. Figs 3, 4, 6 and 7 indicate this for average rate of growth and porosity, Fig. 11 indicates this for β and Figs 13 and 14 indicate this for the saturated width. This effect of p varies as the number of neighbors included in the calculation is changed. The last effect is more noticeable for lower values of p.

A change in the sticking position to accommodate edge-edge and corner-corner type of sticking affects the structure to a large extent. The curves for height and porosity (Figs 4 and 7) and the one for saturated width (Fig. 14) clearly establishes this. An interesting observation is that though the change in sticking position affects the structure drastically, some basic characteristics of the system are not affected. For example, though the saturated width increases with p when edge-edge and corner-corner sticking is allowed as opposed to a face-face only sticking, where it decreases, another parameter given by the ratio of saturated width and the average height growth rate, actually decreases in all the cases. Interestingly, this quantity follows a power law behavior very closely with the attraction parameter, p. Figs 15 and 16 show this for various values of d and sticking positions. It should be noted that for different values of d, as Fig. 15 shows, the power exponent changes its value but the proportionality constant remains same (since at $p = 1$, $W_{sat}/\langle dh/dt \rangle$ has the same value for all values of d). This is not true in the case of Fig. 16 where both the exponent and the proportionality constant changes. Both the figures show that morphological transition takes place at some value of attraction parameter based on the value of d and sticking position. For lower values of p the structures show a rapid change. As Fig. 15 shows, the choice of d strongly affects the value of p beyond which the structural properties saturate. Thus, effects of neighborhood

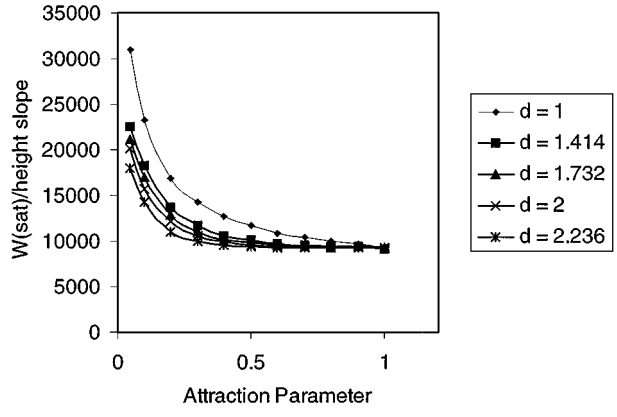

Figure 15 (Saturated width)/(rate of growth of average height) vs. attraction parameter for different values of d.

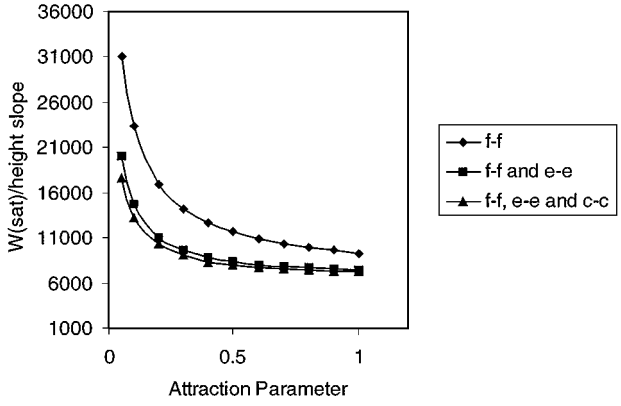

Figure 16 (Saturated width)/(rate of growth of average height) vs. attraction parameter for different sticking positions.

particles are important and have noticeable influence on the structure and morphological transitions.

5. Conclusion

(a) The effects of particle-particle interaction (attraction parameter), interaction length and growth directions (sticking positions) on the deposit structure were studied. The properties of the deposit that were calculated are average height, porosity and roughness.

(b) All the three parameters were found to have effect on the structure to varying degrees.

(c) A new property of the system $W_{\text{sat}}/\langle dh/dt \rangle$ can be defined that can classify all the different types of growth observed here into a single type.

We are working on quantitative aspects of the model to get a clearer picture of the structures and transformations produced by the model.

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References

1. M. J. VOLD, *J. Colloid Interface Sci*. **14** (1959) 168.
2. D. N. SUTHERLAND, *ibid.* **22** (1966) 300.
3. E. S. MACHLIN, in "Materials Science in Microelectronics" (Giro Press, Croton-on-Hudson, NY, 1995) p. 21.
4. H. J. LEAMY, G. H. GILMER and A. G. DIRKS, in "Current Topics in Materials Science," edited by E. Katdis (North-Holland, Amsterdam, 1980) p. 309.
5. P. MEAKIN and R. JULLIEN, *SPIE* **821** (1987) 45.
6. P. MEAKIN, *J. Colloid Interface Sci*. **105** (1985) 240.
7. R. C. BALL and T. A. WITTEN, *Phys. Rev. A* **29** (1984) 2966.
8. F. FAMILY and T. VICSEK, *J. Phys. A* **18** (1985) L75.
9. M. KARDAR, G. PARISI and Y. C. ZHANG, *Phys. Rev. Lett.* **56** (1986) 889.
10. R. BAIOD, D. KESSLER, P. RAMANLAL, L. SANDER and R. SAVIT, *Phys. Rev. A* **38**(7) (1988) 3672.
11. H. F. EL-NASHAR, W. WANG and H. A. CERDEIRA, *Surface Sci.* **391** (1997) 1.
12. J. H. SAUNDERS, J. J. CRISAFULLI and G. H. STICKFORD, ASME Fluids Engineering Conference, June 21–23, 1992, Los Angeles.
13. P. MEAKIN, P. RAMANLAL, L. M. SANDER and R. C. BALL, *Phys. Rev. A* **34** (1986) 5091.
14. F. FAMILY, *Physica A* **168** (1990) 561.

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