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STICKING PROBABILITIES OF FRACTAL AGGREGATES

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Abstract: An attempt was made to experimentally estimate the sticking probabilities of fractal aggregates of fine particles. The experiment was done using MgO fine particles floating in air under normal gravity. The growth in size of the aggregates was monitored by scattered He-Ne laser light. Our preliminary result suggests that the sticking probability decreases with increase in aggregate size. There are, however, still many uncertainties in the method of determining the sticking probabilities. Since this study is still at a developing stage, in this article we provide a general description of the experimental set-up, without giving details of various aspects of the problem.

1. Introduction

It has been known for many years that sticking of fine grains played an important role in the evolution of the early solar system. In the early '70s, for a brief period, it was considered (SAFRONOV, 1969; GOLDREICH and WARD, 1973) that detailed studies of sticking of fine particles were not necessary because gravitational instability of the dust layer near the equatorial plane of the solar system will sooner or later produce planetesimals even if grains do not stick to each other at all. However, by the early '80s, it was realized (WEIDENSCHILLING, 1980; WEIDENSCHILLING and CUZZI, 1993) that the dust layer cannot become sufficiently thin for gravitational instability to occur, if there is turbulent (or convective) motion of the nebula gas. It was also realized (WEIDENSCHILLING, 1980) that even in the absence of such movement of gases, the concentration gradient of the fine grains along the vertical direction of the nebula will produce differential motion of the nebula gas, which will eventually stir up fine grains. Thus, growth of fine particles by some kind of non-gravitational sticking mechanism is an essential process for the formation of planetesimals in the solar nebula.

The size distribution and the structure of aggregates in the nebula as a result of sticking processes are also important parameters which determine the structure of the nebula. This is because the opacity of the nebula, which is mainly determined by the solid particles (as opposed to the gaseous molecules), depends strongly on the aggregate size and structure (LIN and PAPALOIZOU, 1980, 1985; WEIDENSCHILLING, 1984). The smaller the size, and the more porous the structure, the higher the opacity, if the total abundance of solid materials is constant. The opacity is an important parameter which determines the structure of the nebula. If the nebula is very opaque, it could retain heat in the interior and the resultant temperature gradient may drive convective motion of the nebula gas. The typical grain size and the temperature of aggregates in star-forming regions can be estimated from infrared observations. Many recent observations of T-Tauri type stars suggest that aggregates of grains evolve with time (*e.g.*, STOROM and SKRUTSKIE, 1993). The interpretation of such evolution will be incomplete if we do not know how the aggregates stick with each other.

Sticking of fine particles is also an important process with regard to meteorite studies. It is well known that fine grained rims around chondrules, CAIs and other inclusions are the result of accretion processes in the solar nebula (METZLER *et al.*, 1992). But there are many unanswered questions on the accretion processes recorded in chondrites. For instance, we do not have solid answers to the following questions.
1) What is the time scale of the accretion process of the fine grained rims?
2) Did all the fine-grained material accrete as fine grained rims?
3) What is the relationship between the fine-grained rims and the matrix?
4) Did matrix materials accrete as matrix clumps before accreting onto the parent body?
5) Did chondrules accrete into bundles before accreting onto the parent body?
6) How long did the accretion of the parent body take?

Thus, it is important to understand accretion processes of fine grains. The accretion process of two colliding solid grains has been examined theoretically (CHOKSHI *et al.*, 1993). The critical relative velocity for adhesion depends on the grain size and elastic properties and surface energy of the material. However, the process we are interested in is the accretion between aggregates (or between aggregate and a solid grain) consisting of many constituent grains. The degree of freedom of motion of such aggregates are enormous and, at the moment, a theoretical approach to this problem seems helpless. Thus, we decided to conduct an experimental investigation.

The accretion process can be divided into two processes: collision (encounter) and sticking. (Here, we neglect destruction and restructuring of aggregates which could be important at high speed impact.) Nothing happens until two aggregates meet each other. Thus the collision frequency (the rate that a collision happens between particles or aggregates) is an important parameter. It can be roughly estimated using the theory of aerodynamic properties of fine particles, although there remain many problems to be solved.

Once the collision frequency (together with the collision parameters) is known, sticking probabilities can be studied. Sticking probabilities can depend on many parameters, *e.g.* chemical composition, humidity, electric charge, relative velocity, etc.

There are two possible approaches to study sticking of aggregates. One is to observe the outcome (BLUM and MUNCH, 1993) of each collision between two aggregates. If detailed observation can be done, this is a better approach in the sense that what is happening at a collision can be directly observed, even if many observations have to be done. It seems, however, that it is difficult to control the collision parameters in such experiments. Also we do not have a suitable instrument for such observation.

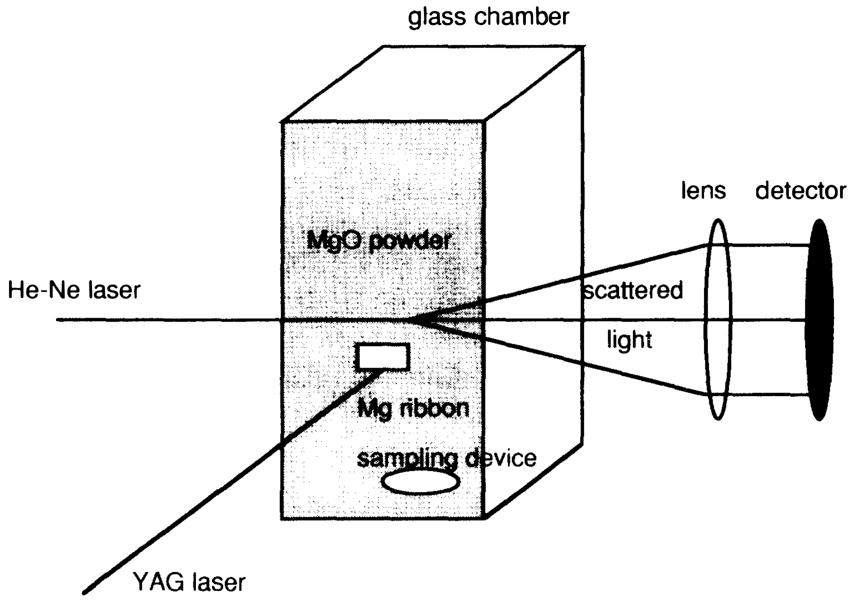
A second approach is to observe the overall growth of aggregates. Changes in the aggregate size distribution with time are observed. In this method we cannot see the details of each collision but can understand directly the time scale of accretion. It is necessary to extract the essence of the sticking process from changes in the size distribution, because we have to extrapolate the laboratory experimental conditions to the solar nebula conditions where gravity, pressure and many other parameters are quite different. Assuming that the collision frequency is well determined, we can (using a simple model) obtain a sticking probability which is averaged over many aggregates. As shown later in this paper, the method of determining the aggregate size distribution is semi-empirical, and the collision frequency determination also has some uncertainty. Nevertheless, we took this second approach, because the experimental conditions are in certain ways similar to the solar nebula conditions; collisions are promoted by convective motion of the gas, which is in turn controlled by the opacity of the aggregates.

2. Experimental Set-up

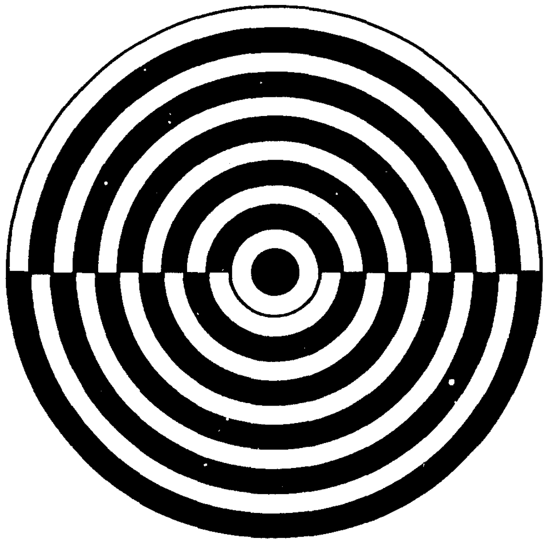
A schematic view of the experimental set-up is shown in Fig. 1a. MgO fine particles are produced by heating (with a YAG laser) Mg metal in the air in a chamber made of glass walls. A 2-mW He-Ne laser with a diameter of 1 cm is shot through the cloud of fine aggregates. The light scattered by the aggregates passes through a lens and is collected on a ring-shaped detector (Tounichi Computer Applications LTD., Fig. 1b). The scattering angle used for the grain size determination is less than 9.3 degrees, which means that we observe so-called forward scattering. The basic principle of the size determination is, the smaller the size, the larger the scattering angle. For instance, light is scattered nearly isotropically by very small size grains (Rayleigh scattering), while scattered light is more focused in the forward direction when the scattering is caused by large grains. This has been exactly demonstrated for solid spheres by Mie theory (Van de Hulst, 1957). For fractal aggregates, as will be shown later, this is an empirical fact with some support from the discrete dipole approximation (DDA) calculations (Draine, 1988) for small aggregates. Since the aggregates have large opacities, a portion of the He-Ne laser light is absorbed in the chamber, producing heat and generating convective air motion. The aggregates mostly follow the air motion, but as the size increases gravitational force becomes more effective and aggregates tend to settle on the chamber floor. There is a sampling device on the bottom of the chamber; the fallen aggregates are collected at several discrete time intervals and observed with an electron microscope for size determination.

3. The Scheme for Estimating Sticking Probabilities

The flow chart in Fig. 2 shows how we determined sticking probabilities of the aggregates. As mentioned briefly above, we do not have an accurate method for size determination of fractal aggregates. We develop a semi-empirical method for the size determination. Then the validity of the method is checked by microscopic observation of fallen aggregates. Since larger aggregates could fall preferentially over small aggregates, the size distribution of fallen aggregates could be (depending on the



a



b

Fig. 1. *(a) A sketch of the experimental apparatus. Aggregates of fine particles of MgO are produced in the chamber by evaporating Mg metal using a YAG laser. A He-Ne laser light scattered by the aggregates is detected by a Si ring-shaped detector placed on the opposite side.*
(b) A sketch of the ring-shaped detector. The dark half-rings are the Si-detecting-elements. The central element (dark circle), which detects unscattered light, is used for alignment of the instrument and also for estimating the opacity of the system. The outermost detector element detects scattered light with a scattering angle of 9.3 degrees.

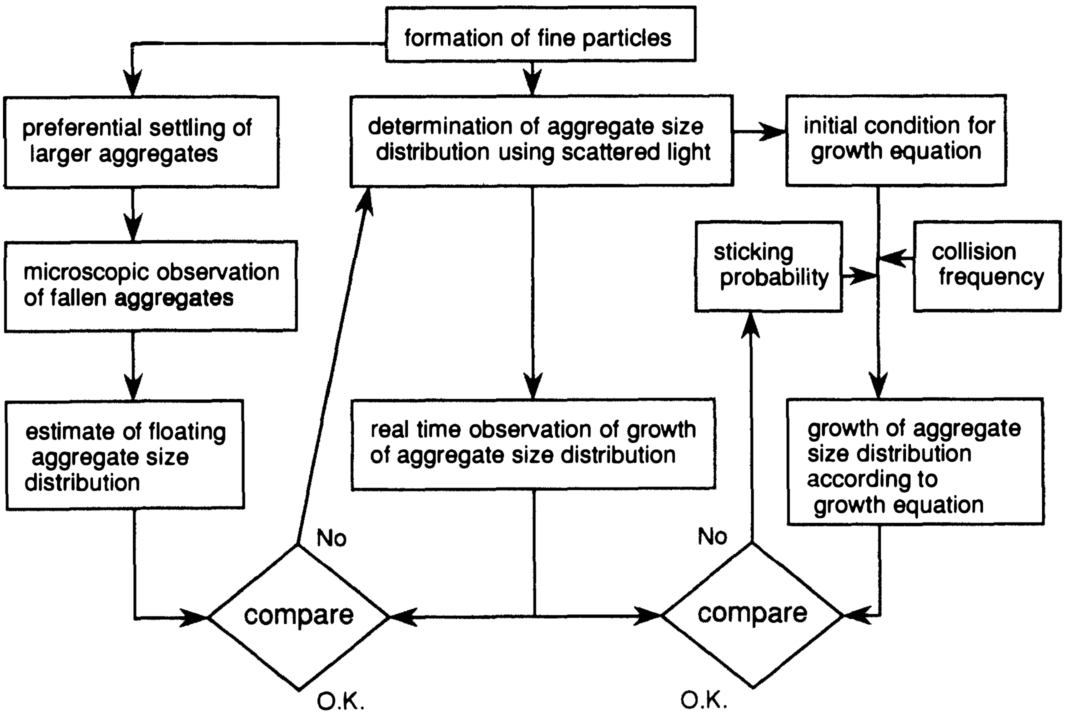


Fig. 2. Flow chart for the study of sticking probability of aggregates.

fractal dimension) biased to the larger size compared with the floating aggregates. It is possible to correct this bias since the hydrodynamic properties of isolated fractal aggregates are fairly well known (MEAKIN *et al.*, 1985). Thus, we are able to estimate the size distribution of floating aggregates at several time intervals. We compare this size distribution with the optically (using scattered light) determined size distribution. If these size distributions are similar, then we conclude that our method of optical size determination is reliable. Otherwise, we make changes to the optical size determination method. Once a practical optical size determination method is established, the next step is to estimate the sticking probability. To do this, we use a growth equation. In this equation there are two main parameters: one is the collision frequency and the other is the sticking probability. As mentioned above, we neglect the aggregate-aggregate hydrodynamic interactions. The estimate of the collision frequency is correct within an order of magnitude. Once this collision frequency is assumed, the only unknown parameter in the growth equation is the sticking probability. Assuming a certain value of the sticking probability, we solve the growth equation numerically and compare the resultant size distribution with the optically determined size distribution. If the results agree well we consider that the sticking probability is correct. Otherwise we change the sticking probability until agreement between the calculated and the observed size distributions is obtained.

4. Inversion Method

The scattered intensity $I(\theta)$ can be expressed as:

$$I(\theta) = \int_{R_{\min}}^{R_{\max}} K(\theta, x)\, n(x)\, \mathrm{d}x, \quad (1)$$

where $x=2\pi\, R_{sca}/\lambda$ is the size parameter, λ is the wave length of the light, θ is the scattering angle and $R_{\min}$ and $R_{\max}$ are the smallest and the largest scattering radii of the aggregates, respectively. The kernel $K(\theta, x)$ is the differential scattering cross section of a single aggregate, and $n(x)$ is the size distribution function which represents the number density of aggregates. The size dependence of the kernel is discussed in the next section. Using the discrete form of eq. (1), the scattering pattern recorded by the detector array can be written as:

$$\boldsymbol{d} = \boldsymbol{K}\cdot\boldsymbol{g} + \varepsilon, \quad (2)$$

where $\boldsymbol{d}$ is the vector of the light intensity detected by l ring-shaped detectors, $\boldsymbol{g}$ is a vector representing the aggregate size distributions in m size classes, $\boldsymbol{K}$ is the $l\times m$ scattering matrix, and ε represents the random measurement error on $\boldsymbol{d}$.

There are various approaches to the inversion of eq. (2) (*e.g.*, summarized in RILEY and AGRAWAL, 1991). In this study, we use the Simplex method (PRESS *et al.*, 1992) which is an iterative procedure with the constraint that the solution vector $\boldsymbol{g}$ is positive ($gi>0$ for any i ($1\le i\le l$)). The Simplex iterative inversion algorithm produces small residuals,

$$\sigma = \sum_{i=1}^{n} \mid d_i^{\text{meas}} - d_i^{\text{calc}} \mid^2, \quad (3)$$

corresponding to near-perfect fits. Here d_i^{meas} represents the measured scattering at detector ring i, d_i^{calc} represents the reconstructed scattering from the estimated size distribution $\boldsymbol{g}$. The solution is rather sensitive to various noises, errors, and residuals of the approximate eq. (2) from the integral eq. (1).

We also examined the Phillips-Twomey method which is a direct matrix inversion with a constraint condition to minimize the norm of the second derivative of the size distribution $\boldsymbol{g}$. The effect of the added smoothness constraint is to reduce resolution but the sensitivity to noise is decreased (CHOW and TIEN, 1976). The least square solution for the eq. (2) is:

$$\boldsymbol{g} = (\boldsymbol{K}^{\mathrm{T}}\cdot\boldsymbol{K} + \gamma\boldsymbol{H})^{-1}\cdot\boldsymbol{K}^{\mathrm{T}}\cdot\boldsymbol{d},$$

where $\boldsymbol{H}$ is a smoothing matrix derived form the second derivative operator, γ is the Lagrangian multiplier that affects the smoothness of the solution. The optimal choice of γ is in practice determined by trial and error. We select the value of γ which minimizes the residual σ in eq. (3) (TANAKA *et al.*, 1982). The direct matrix inversion often presents negative values in the size spectrum if the size range is too broad. Because we reassign the null value to negative values, it gives large σ. Thus we adopt the optimal choice of the minimum and the maximum size class to use in the calculation to minimize σ.

In Fig. 3, the solutions obtained by the Simplex method and the Phillips-Twomey method are compared. These are the solutions for solid spheres (not for fractal

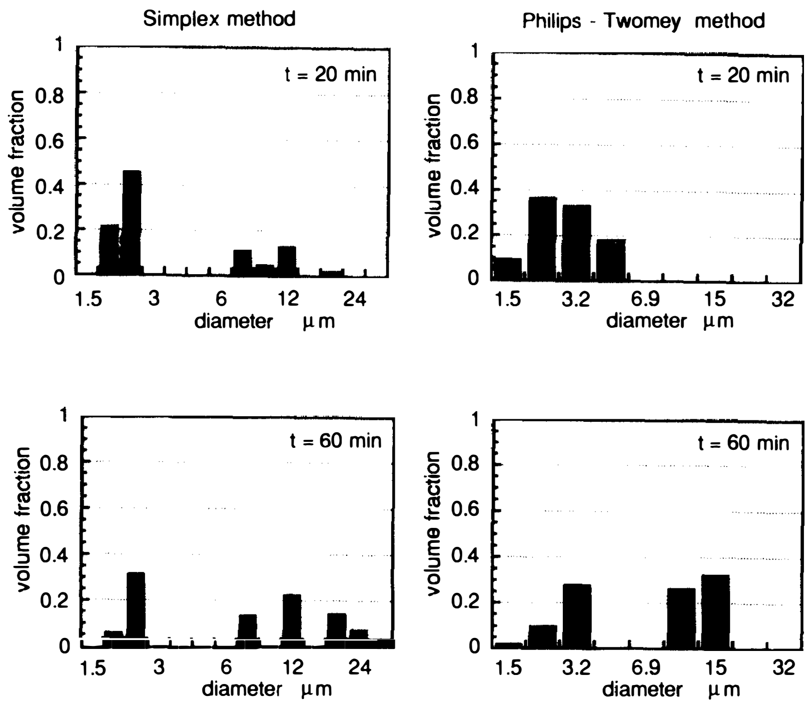


Fig. 3. *Comparison of two inversion methods. Assuming that the light is scattered by solid MgO spheres with a refractive index = 1.735+0i, the size distribution (normalized volume fractions) was estimated by two inversion methods (Simplex (left) and Phillips-Twomey (right)) for time = 20 and 60 min after the start of the measurement.*

aggregates). In the case of t=60 min, the two methods produce fairly similar solutions. In the case of t=20 min, however, the agreement between the two solutions is not very good. At the moment we do not know which method produces a more reliable solution. In the following, we show only the solutions obtained from the Simplex method.

5. Light Scattered by a Fractal Aggregate

At small aggregate sizes, we have a result of DDA which provides accurate information on the scattering properties of aggregates. In principle this DDA can be extended to a larger size, but due to the limit in the computational capacity, at the moment it is not practical to execute the DDA calculation for an aggregate with more than 10000 constituent grains. The DDA for small aggregates suggests that the scattered light is more focused in the forward direction for larger aggregates, similar to the result of the Mie theory for solid spheres. Also we know from our experimental results that the scattering properties change with aggregate size in the size range of 10 to 100 micrometers. Therefore, we decided to extrapolate the DDA result for small aggregates (with up to 512 constituent grains) to larger aggregates using a fractal dimension.

The DDA calculation was performed for aggregates produced by the so-called cluster-cluster aggregation model by computer simulation. The cluster-cluster aggregates have a fractal structure similar to that produced by our experiments. The procedure of the DDA calculation is similar to that explained by KOZASA *et al.* (1992). In Fig. 4, the results of DDA for small fractal aggregates are compared with Mie scattering for spheres with a "scattering radius R_{sca}". Here, the scattering radius is the radius of a sphere for which the scattering pattern (not intensity) in the forward scattering regime is similar to that of the fractal aggregate. It can be seen that such a scattering radius can be chosen for each aggregate as far as the scattering angle is restricted to less than 9.3 degrees. The intensity of the scattered light due to Mie scattering for spheres is not the same as that of DDA for aggregates. To obtain an accurate estimate of the intensity of the scattered light, we use the Maxwell-Garnet theory (BOHREN and HUFFMAN, 1983; HAGE and GREENBERG, 1990). This theory uses average electromagnetic properties of constituent grains for the estimate of scattered light intensity, and is known to provide good results for the estimate of forward-scattered light intensity. To extrapolate the DDA results for small aggregates to much larger size aggregates, we use the fractal dimension. The aggregates produced in our experiments have a fractal structure with a fractal dimension (D) of about 2. We assume that, because of the fractal nature, the scattering radius defined above also has a fractal dimension,

$$N = 2.5(R_{sca} / r_0)^D.$$

Here, N is the number of constituent particles, r_0 is the radius of the constituent particles. The proportional constant 2.5 is determined so that the pattern of the scattered light calculated by Mie theory for a sphere is similar to that calculated by DDA for a fractal aggregate (Fig. 4).

It is noted that in the experimental system we observe mostly single scattering. In most cases, more than 90% of the incident light is transmitted without scattering,

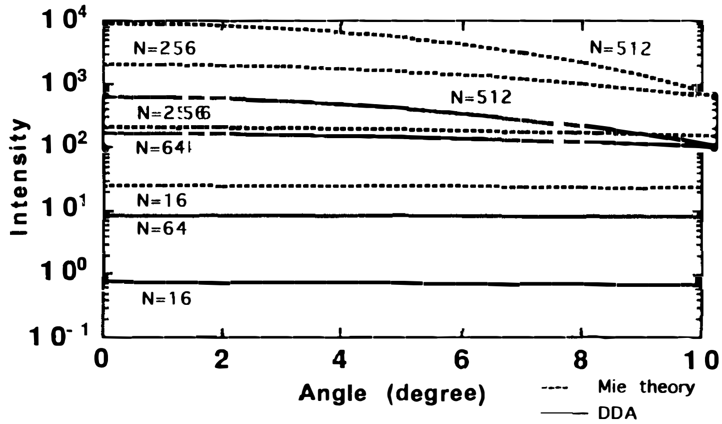


Fig. 4. Angular dependence of scattered light intensity (normalized to the incident light intensity) caused by fractal aggregates calculated by DDA is shown by solid curves for various size aggregates. N is the number of constituent grains in the aggregate. Dotted curves represent Mie scattering by solid spheres with scattering radius corresponding to the fractal aggregates.

indicating that less than 1% of the incident light is multiply scattered. In particular, after the first 10 min of the experiments, due to both growth and settling of aggregates, the transmittance becomes quite close to 1 and the effect of multiple scattering can be neglected.

6. Evaluation of the Optical Sizing Method of Fractal Aggregates

To evaluate the optical sizing method for fractal aggregates described above, the size distributions of aggregates collected on the bottom of the chamber at various times were observed using a scanning electron microscope, and the size distribution was determined. This size distribution could be biased to the larger size compared with floating aggregates because the larger aggregates have a greater tendency to fall

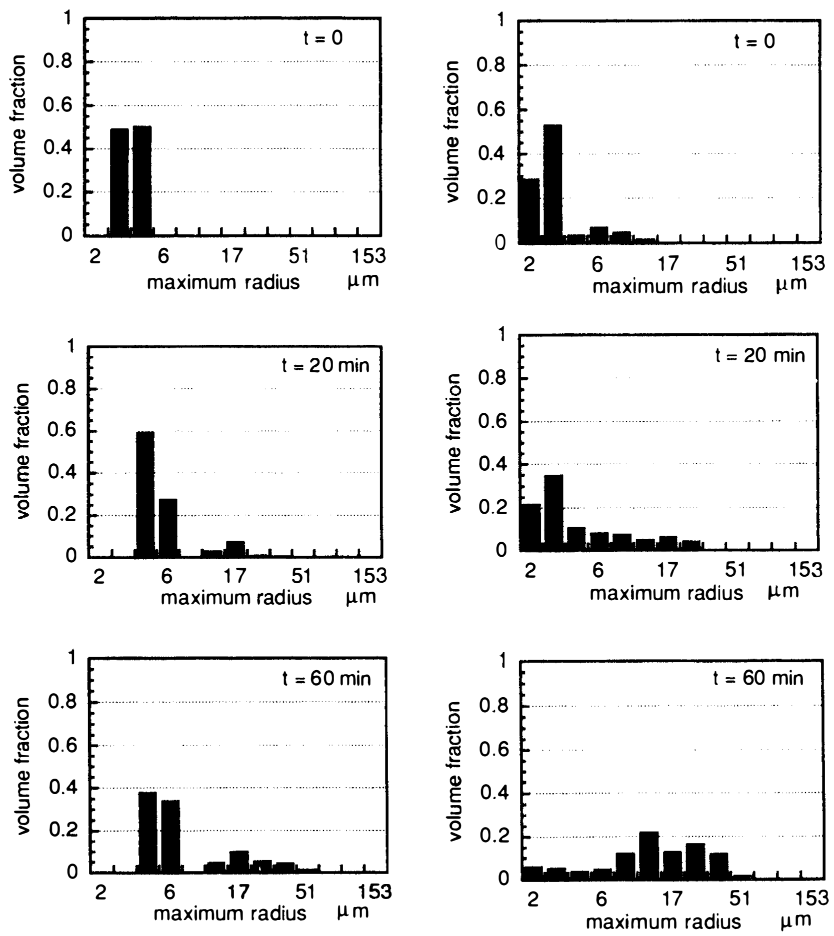


Fig. 5. Growth of MgO aggregates. The evolution is shown from the top to the bottom panel. Each panel shows aggregate size distribution (normalized volume fraction) at 0, 20 and 60 min from the start of the measurement. The left column shows size distributions obtained by the optical sizing method. The right column shows size distributions of aggregates collected on the bottom of the chamber.

than the smaller aggregates. Assuming appropriate friction between an aggregate and air, it is possible to convert the fallen aggregate-size distribution to a floating aggregate size distribution. In the case of aggregates with a fractal dimension=2, it turns out that the terminal velocity is proportional to the square root of the number of constituent particles. This size dependence of the terminal velocity is weaker than that of solid spheres.

The size distribution of aggregates determined by microscopic observation is expressed as a function of the maximum radius of aggregates. The maximum radius is defined by a sphere which encloses an aggregate. This maximum radius has the same fractal dimension as the scattering radius and is about 3 times as large as the scattering radius. Using this relationship between the scattering radius and the maximum radius, the scattering radius determined by the optical sizing method is converted to the maximum radius determined by microscopic observation and compared in Fig. 5. The figures show the normalized volume distribution of each size bin against the maximum radius. Although there is a slight difference at the smallest sizes, the general pattern is fairly similar between the optically determined size distributions and the collected aggregate size distributions. (The size distributions of floating aggregates estimated from the fallen aggregates (not shown) are not much different from the size distributions of fallen aggregates. This is probably a result of the weak size dependence of the terminal velocity.) So we consider that the semi-empirical optical sizing method is not so bad.

7. Simulation of Aggregation

A growth equation,

$$\frac{\mathrm{d}n(r,t)}{\mathrm{d}t} = -n(r,t)\int_0^\infty \beta(r,r')\,n(r',t)\mathrm{d}r + \frac{1}{2}\int_0^r \beta(r-r',r')\,n(r-r',t)\mathrm{d}r' - s(r,t),$$

is used to estimate the sticking probability. Here, r and r' are the radii of aggregates. $\beta(r, r')$ is the collision frequency multiplied by the sticking probability (Φ). $n(r, t)$ is the aggregate size distribution. $s(r, t)$ represents the loss of floating aggregates due to sedimentation onto the bottom of the chamber. It is expressed as v_t/H where H is the height of the chamber (40 cm) and v_t is the terminal velocity of aggregates.

It is not easy to estimate the collision frequency between fractal aggregates under atmospheric pressure. The collision frequency when the grain size is much smaller than the mean free path of gas molecules (the solar nebula condition) can be estimated with fair certainty, since the effect of the gas on the trajectories of grains can be neglected at the time of collision. In our experiment, growth of aggregates floating in air is observed under 1 atmospheric pressure, because under smaller pressure (and normal gravity) aggregates fall quickly before accretion takes place. In the case of collisions between two solid spheres in air, the collision frequency is accurately known (ZHANG and DAVIS, 1991). But, it is not well known what the hydrodynamic effect of the gas may be when two aggregates are close together under 1 atmospheric pressure. In this study we assume that the hydrodynamic interaction between two close aggregates can be neglected because of their open fluffy structures.

The validity of this assumption will be a subject of our future study.

Brownian motion and differential settling due to gravity are important collision mechanisms. Details on this subject have been described in FUCHS (1964). In short, Brownian motion is more effective for small size aggregates and the differential settling predominates at larger sizes. The collision rate due to differential settling is zero if the sizes of the two aggregates are equal. This causes a problem in numerical simulation because many aggregates are put into the same size-bin, especially in the early stage of accretion, and if taken at face value they do not collide at all with each other. In reality, there is a substantial difference in size even for grains in the same size-bin, and they could collide with each other. (Shear flow is another mechanism which makes equal size aggregates collide with each other.) Thus, we assume that the growth rate parameter $\beta(r, r')$ has the following form.

$$\beta(r, r') = A\,\Phi\,(R_{\max,\,i} + R_{\max,\,j})^3.$$

This is a shear flow type collision function but is a fair approximation of the Brownian+differential settling collision function, with a constant $A=13.3$.

Aggregate motion is assumed to be similar to the motion of a solid sphere with a radius which is called the hydrodynamic radius (R_h, MEAKIN *et al.*, 1985). It is assumed that the R_h has the same fractal dimension, D, as the scattering radius, and is expressed by:

$$N = 0.5(R_h / r_o)^{D}.$$

Here, N is the number of constituent grains in an aggregate and r_o is the size of the constituent grains. The choice of the constant 0.5 is rather arbitrary but cannot be very wrong because the hydrodynamic radius is not very different from the maximum radius. The loss term $s(r, t)$ in the growth equation is calculated using this hydrodynamic radius and the Stokes law.

Definition of the collision cross section between fractal aggregates is not obvious. The number density of constituent grains near the periphery of an aggregate is rather small. But it is assumed that, considering that aggregates are rotating, a collision occurs when the distance between two aggregates becomes less than the sum of the maximum radii of two aggregates. The sticking probability is assumed to be either constant or a function of aggregate size. With these assumptions we trace the evolution of the growth equation numerically, using the size distribution based on the first optical observation as an initial condition.

8. Results and Discussion

In Fig. 6, two examples of the numerically obtained aggregate size distributions and experimentally obtained size distributions are compared at several time steps. In one example, the sticking probability is assumed to be one, *i.e.* aggregates stick at every collision. It can be seen that in this case the numerically obtained size distributions have rather different shape when compared with the experimentally determined size distributions. This seems to be the result of quick growth of larger aggregates in the numerical simulation. Another example in which the sticking probability is assumed to be inversely proportional to the number of constituent grains was examined. In this case, it can be seen that the rough qualitative agreement

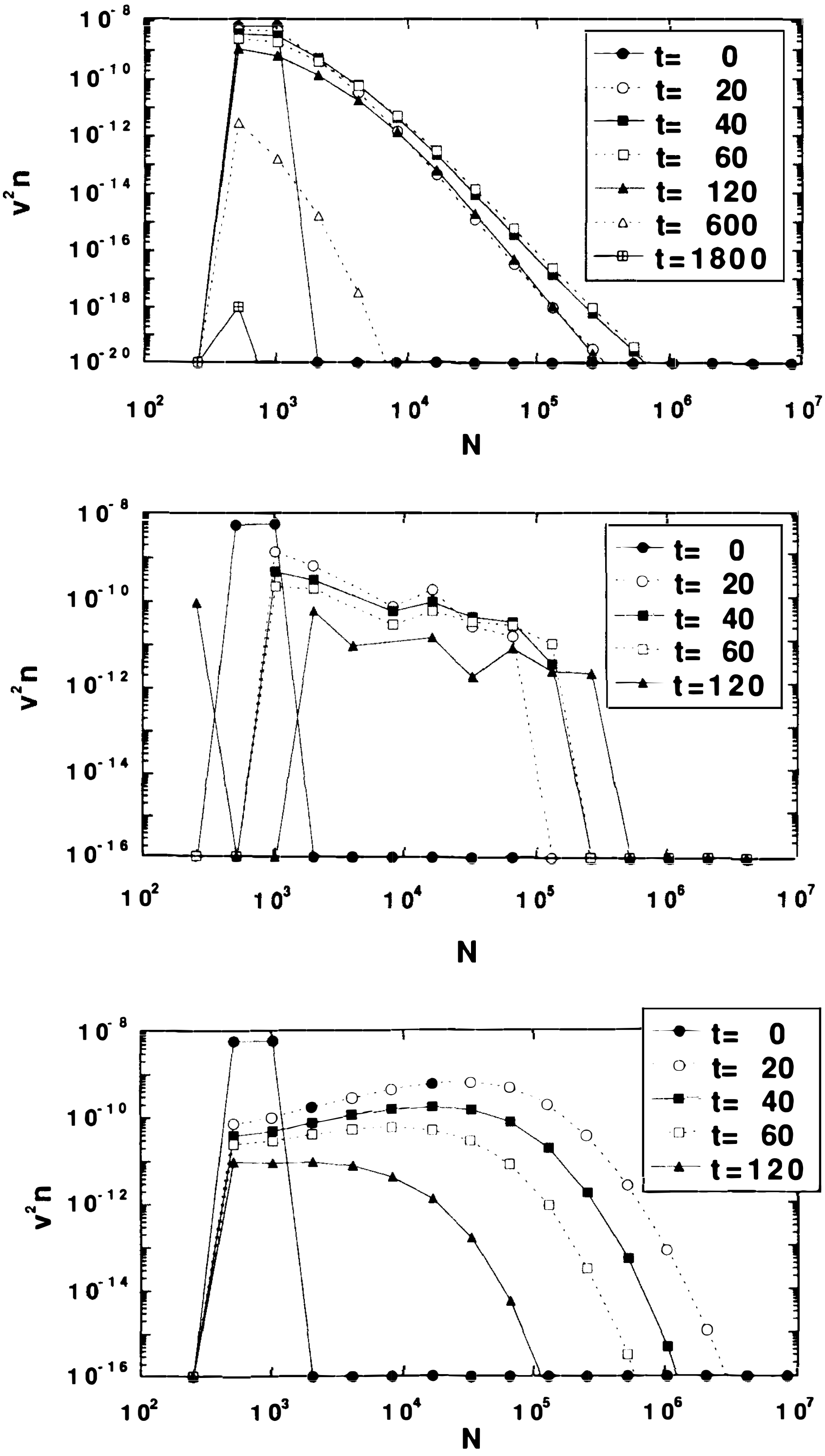


Fig. 6. *Comparison of aggregate size distributions. Vertical axis shows volume density. t is time in minutes after the start of the first measurement. The top panel shows calculated MgO aggregate size distributions assuming that the sticking probability is 1. The center panel shows MgO aggregate size distributions determined by the optical sizing method. The bottom panel shows calculated aggregate size distributions assuming that the sticking probability is inversely proportional to the number of constituent grains.*

with the experimentally obtained size distribution, *i.e.* a relatively flat distribution in the intermediate size range, is obtained, although a very large proportional constant$=10^5$ is required. There may be two reasons to expect that the sticking probability may be small for large aggregates. First, large aggregates tend to have large relative velocity at collisions. Second, a large fraction of collisions between large aggregates are grazing collisions. Since fractal aggregates have only a small number of constituent particles on the periphery, the momentum and the angular momentum of collision may be too large to be supported by the adhesive force between the small number of peripheral constituent particles. There are a couple of reasons that we have a better fit with a large proportinal constant. First, the size of the actual MgO constituent particles is not uniform but ranges from 0.05 micrometer up to a few micrometers. Aggregates with large constituent particles could collide more often with other aggregates. Attractive force, such as electric charge may also work to enhance the accretion rate. There remains a possiblity, however, that our estimate of the collision frequency is grossly underestimated.

We do not yet attach much significance to our results. As noted above, many uncertain assumptions are used to derive the sticking probability. These assumptions have to be examined more rigorously and improved in order to have confidence in the results. Considering the fact that accretion has not yet been observed in another approach (BLUM and MUNCH, 1993) to this problem, however, the present result that fractal aggregates grow to a size of several tens of microns with a time scale of about 1 hour is an important step forward to understanding the accretion of fine grain aggregates.

Acknowledgments

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