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Microgravity studies of aggregation in particulate clouds

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[1] Aggregation in clouds of submillimeter quartz and volcanic ash particles was studied in microgravity. Particle clouds generated by pulses of air immediately formed electrostatic filamentary aggregates upon cessation of air turbulence. Manual agitation of experiment chambers produced cm-size loose grain clusters which voraciously scavenged free-floating material in their vicinity. A dipole model accounts for these observations. Experimental results have ramifications for the behavior of natural cloud systems and primary accretion of solids in the early solar nebula.
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1. Introduction

[2] Ground-based and KC-135 reduced-gravity aircraft experiments with clouds of basaltic dust and quartz sand [*Marshall et al.*, 1981, 2001] have shown that turbulently disturbed (tribocharged) particulate systems very rapidly aggregate into filaments upon cessation of turbulence, or they persist as large cohesive masses if dispersion is inefficient. Filamentary aggregation in particulate clouds is commonly observed and has been attributed to a variety of causes including bipolar grain interactions [*Whitby and Liu*, 1966], permanent dipoles on individual grains (D. Bieschel and A. Winkel (1936) as discussed by *Fuchs* [1964]) short range dipolar induction [*Nielsen and Hill*, 1980], electrical field effects [*Sharma*, 1992], and complex interactions of monopoles, electrostatic shielding, and ballistic forcing of particle contact [*Huang and Kushner*, 1997].

[3] We report on Space Shuttle experiments conducted as a follow-up to our ground-based and KC-135 tests. Results confirm the propensity of agitated particulate clouds to "spontaneously" spawn populations of electrostatic filamentary aggregates and, under certain circumstances, to evolve large, loosely-packed electrostatic clusters.

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2. Experiments

[4] Experiments were conducted on the First and Second U.S. Microgravity Laboratories (USML-1, 2) aboard Space Shuttle Columbia; Coulombic forces in aggregation were gravitationally unmasked by elimination of particle self-weight and sedimentation dynamics, and by facilitating protracted particle suspension in 10^{-5} g. All experiments used sand-size material, chosen in preference to dust because it is relatively easy to disperse, and individual grains could be imaged with inexpensive cameras.

[5] The flight apparatus (Figure 1) consisted of eight miniature cloud chambers of 125 cc. Each was individually plugged into a pump base. Modules were self-contained experiments, loaded prior to launch with several grams of material. The pump unit was a hand-cranked piston device for generating 100 cc of 82 kPa (12 psig) compressed air released as a jet through the base of an attached module. The jet forcefully dispersed grains around the inside of the module with the air escaping through chamber-wall screens -dispersion caused triboelectrostatic charging of test materials. Particles were imaged as silhouettes by diffused back lighting. All experiments were recorded on videotape. Cloud densities immediately after dispersion were estimated to be several tens of grains per cc for 0.4 mm material, for example (with initial module load of 2 gm). Cloud densities implied that 50–75% of the material stuck to the chamber walls, corresponding to visual estimates made by the astronauts. Dispersing air motion damped out after ~30 seconds, and grains were then left undisturbed for up to 30 minutes. In some experiments, astronauts mechanically dispersed the grains by rocking the modules.

[6] The effect of grain size on aggregation was tested in ML-1 with three diameters of quartz grains (Table 1). Material type was tested in ML-2 using non-conducting, homogeneous quartz and heterogeneous volcanic ash, with conducting copper grains as a control. A copper-quartz combination in ML-2 tested a conductor-insulator mixture. Module 05 of ML-2 had angular quartz grains for comparison with rounded quartz in other modules. Module 06 of ML-2 had four 5 mm volcanic pebbles added to the volcanic ash to determine if they would act as platforms for aggregate growth. The role of initial cloud density was tested in ML-1 with two different grain loads for quartz, and in ML-2 with three different grain loads for both quartz and ash. To test interior wall effects, insulating anodized aluminum walls in ML-1 were changed to conductive alodined surfa-

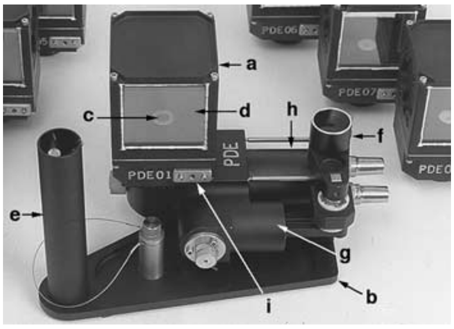

Figure 1. USML flight hardware showing pump unit and attached module. For scale; module windows ~5 × 5 cm. a: Plug-in module, b: Pump unit, c: Air-jet orifice, d: Module front window, e: Grip, f: Crank handle, g: Pump piston, h: Compressed-air reservoir, i: Air-valve trigger.

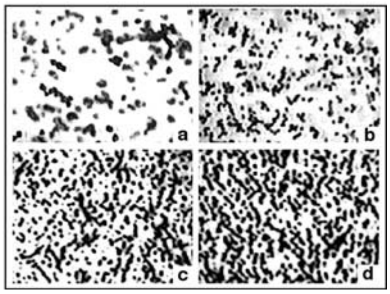

Figure 2. Filamentary aggregates in microgravity. See Table 1 for parameters. a: Module 1, ML-1, b: Module 03, ML-1, c: Module 03, ML-2, d: Module 2, ML-2. Images have field widths of ~2.5 cm.

ces in ML-2. Module 04 in ML-2 was identical to 05 in ML-1 as a control.

3. Results

3.1. Aggregation of Nominally Monodispersed Grains

[7] As soon as grain motion damped out after dispersion, both quartz and volcanic grains instantly formed populations of filamentary aggregates. The same electrostatic forces creating aggregates also contributed to damping of ballistic motion—an effect of Coulombic friction [*Marshall*, 1998]. Filamentary aggregates typically consisted of single chains of particles ranging in length from simple couplets to strands with tens of grains (Figure 2). Statistical counts of whole aggregate fields (not possible for all materials) established that filaments of 0.4 mm quartz had a mean length of five grains. In both dilute and dense grain populations, filaments adopted straight, kinked, wavy, and bifurcating forms. Denser clouds tended to produce longer filaments. Assessment of aggregation for 0.1 mm quartz grains and from copper and the copper-quartz mixture were inconclusive owing to dispersion and/or illumination problems.

[8] Aggregation rates and styles were not obviously different between angular and rounded quartz. Nor were they noticeably different between volcanic ash and quartz. Wall differences between ML-1 and ML-2 had no obvious effect on aggregation, nor on the amount of material adhering to the walls. In the experiment with large pebbles mixed with sand, pebble surfaces acted as attractors for aggregates and free grains only when there was gentle (induced) relative motion of the materials involved in the interaction. Within the constraints of the test matrix, filamentary aggregates were universal–their occurrence being independent of grain size, shape, and composition, cloud density, ballistic energy of grains, and experiment duration.

[9] Virtually all aggregation occurred within the first few minutes after dispersion, with only a few grains repositioning themselves during extended test durations of up to 20–30 minutes. Cessation of aggregation is considered to have resulted from three factors: 1) decay of electrostatic charges after the initial impulse of tribocharging, 2) depletion of grain numbers in areas between aggregates, and 3) reduction in Coulombic interactions owing to a transition from disorganized forces (during dispersion) to organized forces (after aggregation). Despite close proximity of aggregates in some ML-2 experiments, we did not observe them combining into larger clusters. Aggregates had quasi-stable arrangements of materials and forces that appeared inert on the timescales of the experiments.

[10] Repulsion between grains was not observed in any experiments. However, repulsion is difficult to substantiate from observations of grain motion because it does not have obvious end products such as grain-grain sticking (aggregation) observed between attracting materials.

3.2. Behavior of Cluster Aggregates

[11] When modules were agitated by the astronaut, material detached from the walls as large, but intact, clumps ranging in size from clusters of tens of grains to clusters of tens of thousands. In some cases, clusters showed no tendency to attract other free-floating material, even though grains within them were evidently attracting one another. In

Table 1. Module Contents[a]

USML-1				USML-2			
Module	Material	Grain Size (mm)	Weight (g)	Module	Material	Grain Size (mm)	Weight (g)
01	q	0.8	3	01	v	0.4	3
02	q	0.1	3	02	v	0.4	3
03	q	0.4	3	03	q	0.4	3
04	q	0.8	2	04	q	0.4	2
05	q	0.4	2	05	aq	0.4	2
06	q	0.8	2	06	v + p	0.4 + 5.0	3
07	q	0.1	2	07	c	0.4	10
08	q	0.1	2	08	c + q	0.4	5.0 + 1.5

[a]q = quartz (rounded alluvial sand), v = volcanics (Mt Shasta ash), v + p = volcanic grains mixed with four 5 mm pebbles, aq = angular quartz (crushed crystals), c = copper (elongate filings, grain diameter nominal).

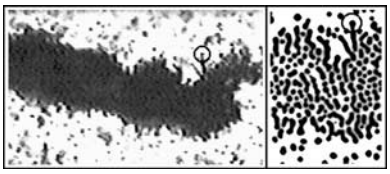

Figure 3. Cluster aggregate of 0.4 mm quartz grains showing surface growth of filaments. Photo field is ~2 cm wide. Image processing on right (magnified) shows coaxial alignment of filamentary fabric on (and within) a section of the aggregate. Circle provides reference location.

other cases, clusters acted as powerful electrostatic attractors, rapidly scavenging free-floating grains and filaments from all directions. No grains were observed to be repelled by these clumps of material. In one particular case observed, the electrical field associated with the cluster scavenged material radially from several centimeters away, causing "infalling" grains to attain speeds up to 1 cm.s^{-1} (see discussion). Grains attracted to these cluster aggregates formed filaments that grew normal to the aggregate surface (like a hairbrush). In the example in Figure 3, gentle sideways movement of the module by the astronaut caused a cluster to drift from one side of the module to the other within ~10 seconds. The aggregate began with a relatively smooth surface but by the time it reached the middle of the module, it was bristling with filaments created from material swept up in its path.

[12] Cluster aggregates were shown to be composed of coaxially-aligned filaments when video images were enhanced with Adobe Photoshop©, as seen in Figure 3. This explains their attractive power, which is analogous to that of a magnet whose strength depends on magnetic domain alignment. Cluster aggregates that did not attract surrounding material were lacking this alignment fabric.

4. Discussion

4.1. Grain Interaction Model

[13] Grains experienced triboelectrification during air-pulse dispersion, or when modules were agitated by the astronaut. Collision of grains led to charge exchange at contact points and created a random distribution of both positive and negative charge spots on each grain surface [*Cross*, 1987; *Abrahamson and Marshall*, 2002]. An imbalance of opposite charges produces a monopole (net charge), while irregular charge distribution produces a dipole. We suggest that dipoles are responsible for filamentary structures. As grains approach one another, they rotate so that their dipole axes are mutually attractive and in coaxial alignment. Other approaching grains find the end of a filament to be more attractive than positions along its axis owing to the configuration of electric field lines around the aggregate.

[14] Particle chains could also be a result of alternate stacking of positive and negative monopoles (this also creates dipoles). In order for this to occur, grains of like material (e.g., quartz) must be able to charge either positive or negative. It follows that any single grain can acquire charges of both sign because charge spots are electrically independent of one another on dielectric surfaces. Most frictionally charged surfaces have both positive and negative charging, with one charge dominating [*Cross*, 1987]. Thus, any argument for bipolar stacking inherently accounts for dipoles. With the volcanic materials, different grain compositions could have acquired different charge signs, but this does not negate mixed charging on any single grain composition.

[15] Monopole-monopole (M-M) attraction varies as $1/r^2$ and dipole-dipole (D-D) attraction varies as $1/r^4$, where r is the separation distance between charges. Dipole forces (F_D) therefore tend to be stronger at short range, but rapidly decline with distance and become weaker than monopoles (F_M). At relatively large intergranular spacings where F_M dominates, grains attract or repel one another, but at close range, inside a critical separation distance where $F_D = F_M$, grains always attract one another by rotation of their dipole axes, regardless of any monopole repulsion. While M-M, D-D, and M-D attractions can explain filaments, only D-D attractions account for large aggregates attracting but not repelling grains in their vicinity. If large aggregates had been of opposite charge to grains attracted to them, they would have been neutralized by accretion. There is no reason to assume that grains attracted to cluster aggregates were electrostatically different from grains in other experiments, hence we deduce that dipoles were an important, if not dominant control of grain aggregation in all modules.

4.2. Calculation of Dipole Forces

[16] One of the large clumps of 0.4 mm quartz grains was surrounded by a ballistically quiescent dispersed grain population, enabling dipole forces on typical grains to be estimated, since grains could be tracked from a zero-velocity position. Velocities of grains attracted to the aggregate were measured from videotapes of the experiment. We modeled electrostatic force (F_E) between the large clump and an individual monomer as a dipole-dipole force: $F_E = 6D_1D_2/4\pi\varepsilon_o r^4$ where D_1 and D_2 are the dipole moments of the clump and a monomer, respectively, and $4\pi\varepsilon_o$ is a constant (numerically equal to 1 in the esu system) with units of esu^2 s^2 g^{-1}cm^{-3}. Either assuming the grains to be moving at terminal velocity, or in free fall, we obtain an acceleration of 0.04 cm.s^{-2}, and find the product of the grain and aggregate dipole moments $D_1D_2 = 10^{-5}$ to 10^{-4} esu.cm. It must be that $D_1 \gg D_2$, since the grains were attracted much more strongly to the cluster than to each other.

[17] As a first estimate, we assume that $D_1 = f N D_2$, i.e., that the large aggregate is composed of N grains identical to the isolated ones, with a fraction f having their dipole moments all aligned (Figure 3). For the aggregate under study, we roughly estimated $N = 10^4$ and $f = 0.1$, although clearly there is uncertainty in these values. We thus obtain $D_2 = 1\text{–}2 \times 10^{-4}$ esu-cm, or a separated charge of about 10^6 elementary charges. The aggregate would then have $D_1 = 0.1$ esu-cm, or about 10^8 elementary charges. For either the monomers or the clump, the surface charge density was only 5–10% of the Gaussian limit [*Cross*, 1987].

5. Implications for Particulate Systems

[18] It was observed that even brief turbulent suspension of particles leads to sufficient tribocharging to induce rapid

aggregation, that aggregation can produce nearly an order of magnitude increase in effective particle size, and that dipoles provide a potent (always) attractive force in addition to M-M interactions in a cloud.

5.1. Volcanic Clouds

[19] These factors should enhance aggregation in volcanic eruption plumes where cloud densities are similar to, or even higher, than those in USML. Electrostatic processes have been implicated in the growth of various aggregate species such as accretionary lapilli, small (~mm) tightly bound spheres precipitated from eruption clouds, and fist-size, aggregate "hail" reported during the Mount St. Helens eruption [*Sparks et al.*, 1997; *Hobbs et al.*, 1981]. Filaments could provide nucleii for such structures, with dipoles contributing to both their formation and cohesive strength.

[20] By analogy with the microgravity experiments, a momentary waning of turbulence within pockets of an eruption plume will lead to localized, spontaneous production of aggregates. Simultaneously, the quiescence will permit gravitational sedimentation of the aggregated material. In particular, peripheral parts of an eruption column should experience more frequent aggregation opportunities. We also envisage regions of the plume where there is a dynamic equilibrium between gravitational forces driving precipitation of aggregated material and buoyancy forces from ascending gas flow, a situation found in thunderstorms where hail stones are accumulated prior to sudden precipitation. Volcanic aggregate "hail" composed of clusters similar to those in Figure 3 could grow by scavenging particles and small filaments streamed past them while they are suspended in the flow. As an eruption wanes, gas content reduces and/or erosion widens the vent, and the eruption column can no longer be sustained –column collapse ensues, leading to pyroclastic flows and surges [*Chester*, 1993]. We suggest that gravitational collapse can be enhanced, perhaps even driven by bursts of aggregate hail [*Marshall et al.*, 1998]. Dipole-driven aggregation, cohesion, and Coulombic friction could be important contributors to the sedimentation dynamics of dry eruption plumes and clouds.

5.2. Planetary Dust Palls

[21] In contrast to volcanic eruption plumes, aeolian dust clouds on both earth and Mars are very dilute systems where we might expect M-M interactions to dominate owing to large inter-particle separations. Any net charge on a cloud as a whole acts as a dispersive force, helping to maintain these separations. During sedimentation, differential particle settling rates might increase the probability of particle encounters involving D-D interactions if settling velocities force particles through M-M repulsive barriers. Particulate clouds are also generated by meteorite impact. Owing to the violent nature of these events, strong electrostatic charging would be expected on the lofted material. Close to the impact site, D-D interactions may dominate where pall densities are high—similar to the volcanic case, but in distal regions, M-M interactions may dominate – similar to the aeolian case.

5.3. Protoplanetary Nebula

[22] USML results also have a bearing on the long-standing debate in astrophysics concerning sticking and coalescence of protoplanetary particles in the nebula [*Marshall and Cuzzi*, 2001]. A binding energy of $3 - 12 \times 10^{-4}$ ergs between two 0.4 mm monomers calculated from our observed dipole forces is 1000–4000 times larger than the critical van der Waals binding energy, previously assumed to dominate sticking of silicate grains, typically 10^{-7} ergs for 0.4 mm grains [*Dominik and Tielens*, 1997, Figure 4]. Experiments with micron-size grains [*Poppe et al.*, 2000] indicate sticking at energies 100 times larger than predicted by van der Waals forces [*Dominik and Tielens*, 1997], and it is conceivable that electrostatic forces contributed to these values. Replacing the van der Waals binding energy of *Dominik and Tielens* [1997] with our dipole binding energy and assuming that ~100 contacts participate in a collision of a particle with an aggregate, this results in predicted stability for chondrule aggregates up to encounter velocities of 15 $cm.s^{-1}$, consistent with our observations.

6. Conclusions

[23] Colliding grains in a turbulent cloud acquire electrostatic dipoles which can override monopole forces at short range, leading to the formation of filamentary aggregates and enhanced aggregation rates. Dipoles might play a significant role in particulate cloud behavior and in the more general area of particle adhesion and cohesion. Experimental results suggest that they may be ubiquitous in tribocharged particulate systems involving dielectric materials.

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References

Abrahamson, J., and J. R. Marshall (2002), Permanent electric dipoles on gas-suspended particles and the production of filamentary aggregates, *J. Electrostatics*, *55*, 43–63.

Chester, D. (1993), *Volcanoes and Society*, 117 pp., Edward Arnold, London.

Cross, J. A. (1987), *Electrostatics*, Adam Hilger, Bristol, U.K.

Dominik, C., and X. Tielens (1997), The physics of dust coagulation and the structure of dust aggregates in space, *Astrophys. J.*, *480*, 647–673.

Fuchs, N. A. (1964), *The Mechanics of Aerosols*, 313 pp., Elsevier, New York.

Hobbs, P. V., L. F. Radke, M. W. Eltgroth, and D. A. Hegg (1981), Airborne studies of the emissions from the volcanic eruptions of Mount St. Helens, *Science*, *211*, 816–818.

Huang, F. Y., and M. J. Kushner (1997), Shapes of agglomerates in plasma etching reactors, *J. Appl. Phys.*, *81*(9), 5960–5965.

Marshall, J. R. (1998), "Coulombic viscosity" in granular materials: Planetary and astrophysical implications, *Lunar Planet. Sci.*, *XXIX*, 1135–1136.

Marshall, J. R., and J. Cuzzi (2001), Electrostatic enhancement of coagulation in protoplanetary nebulae, *Lunar Planet. Sci.* [CD-ROM], *XXXII*, Abstract 1962.

Marshall, J. R., D. H. Krinsley, and R. Greeley (1981), An experimental study of the behavior of electrostatically-charged fine particles in atmospheric suspension, *NASA Tech. Memo.*, *84211*, 208–210.

Marshall, J. R., F. Freund, T. Sauke, and M. Freund (1998), Catastrophic collapse of particulate clouds, *NASA Tech. Memo.*, *1998-208697*(35), 579–592.

Marshall, J. R., T. Sauke, and W. Farrell (2001), Electrostatics of granular materials, preliminary science requirements document, NASA Glenn Res. Cent., Cleveland, Ohio.

Nielsen, K. A., and J. C. Hill (1980), Particle chain formation in aerosol filtration with electrical forces, *AIChE J.*, *26*(4), 678–686.

Sharma, A. (1992), Dendritic aggregation of metal hydrides and oxides in a laser-produced aerosol medium, *Phys. Rev. A*, *45*(6), 4184–4187.

Poppe, T., J. Blum, and T. Henning (2000), Experiments on collisional grain charging of micron-sized preplanetary dust, *Astrophys J.*, *533*, 472–480.

Sparks, R. S. J., M. I. Bursik, S. N. Carey, J. S. Gilbert, L. S. Glaze, H. Sigurdsson, and A. W. Woods (1997), *Volcanic Plumes*, 457 pp., John Wiley, Hoboken, N. J.

Whitby, K. T., and Y. H. Liu (1966), The electrical behaviour of aerosols, in *Aerosol Science*, edited by C. N. Davies, Elsevier, New York.

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