Structure and Mechanical Properties of High-Porosity Macroscopic Agglomerates Formed by Random Ballistic Deposition

Jürgen Blum* and Rainer Schräpler
Institut für Geophysik und Extraterrestrische Physik, Technische Universität Braunschweig, Mendelssohnstrasse 3, 38106 Braunschweig, Germany
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We present experimental results on the mechanical properties of macroscopic agglomerates formed by ballistic hit-and-stick deposition. The agglomerates, produced with a new experimental method, consist of monodisperse SiO_2 spheres with 1.5 μm diameter and have a volume filling factor of $\phi = 0.15$, matching very closely the theoretical value for random ballistic deposition. They are mechanically stable against unidirectional compression of up to 500 Pa. For pressures above that value, the volume filling factor increases to a maximum of $\phi = 0.33$ for pressures above 10^5 Pa. The tensile strength of slightly compressed samples ($\phi = 0.2$) is 1000 Pa.

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The range of packing densities of equal-sized spheres is a long-standing problem. Most publications deal with the discussion of the densest packing of spheres, mainly with random close packing (RCP). The volume filling factor ϕ, defined as the fraction of the volume filled by spheres, in RCP is close to $\phi = 0.64$ (see, e.g., the discussion in Ref. [1]). In this Letter, we present experimental results on our work on the formation of planetesimals, the kilometer-sized precursors to the planets of our Solar System [2] which are, due to the absence of high-velocity collisions and considerable gravitational forces, much less compacted than RCP. In the phase preceding the formation of these first bodies with sufficient gravitational attraction, it is assumed that the initially micrometer-sized dust grains agglomerated due to low-velocity collisions and attractive interparticle forces. The major objective of our work is to understand the so-called runaway agglomeration process in which a few large dust agglomerates grow much faster than the majority of the dust grains by an irreversible collection of the generally much smaller dust particles encountered (see Ref. [2] for details). Therefore, we are interested in the idealized ballistic deposition processes in which a target agglomerate consisting of a large number of monomers grows by accumulation of single monomer particles that hit the target agglomerate on ballistic trajectories. For sticking probabilities of unity and for random directions of the impinging particles, the forming agglomerates are quasi-spherical in shape and have volume filling factors of $\phi = 0.15$ [3,4]. When monomer particles are deposited on a semi-infinite target agglomerate at random positions but from a single direction (in this case perpendicular to the target surface), the process is referred to as random ballistic deposition (RBD). Watson *et al.* [5] report on RBD simulations with 10^6 particles and find $\phi = 0.1469 \pm 0.0004$, which is in excellent agreement with our own Monte Carlo simulations with also 10^6 particles [see Fig. 2(c)].

For the experimental realization of RBD and the comparison to numerical models, it is required to have (i) monodisperse, spherical particles, (ii) attractive surface forces F_{stick} between the particles that exceed the particle weight considerably [6], i.e., $F_{stick} \gtrsim 10^6 m_0 g$, where m_0 and g are the particle mass and the gravitational acceleration, respectively, and (iii) a hit-and-stick behavior in any collision between a particle and the target agglomerate. In Ref. [7] it was shown that for spherical particles a well-defined sticking threshold velocity v_{stick} exists below which particles always and above which particles never stick. Particle rolling in collisions is prevented for impact velocities $v_{im} < v_{roll} = \sqrt{(10E_{roll})/m_0}$ [8,9]. Here, E_{roll} is the energy required to roll one monomer grain over a quarter of its circumference along the surface of another monomer grain [8]. E_{roll} can be obtained by measuring the rolling-friction force [10]. Thus, hit-and-stick collisions occur whenever particles collide with the target agglomerate with impact velocities $v_{im} < v_{hs} = \min(v_{roll}, v_{stick})$.

Close-to-perfect monodisperse spherical particles are commercially available. Because of the applicability to preplanetary processes, we chose the sicastar SiO_2 particles from *micromod Partikeltechnologie GmbH* whose characteristics are detailed in Table I. The particles have smooth surfaces [10] and the ratio of adhesion force and gravitational force for these particles is $\chi \sim 2 \times 10^6$ and therefore even beyond the limit of the dynamical simulations in Ref. [6] where a beginning saturation of the volume filling factor is observed at $\chi > 10^5$ for sedimenting particles.

The experimental setup (see Fig. 1) consists of a cogwheel deagglomerator [12] with which the powder sample is disintegrated into its monomer grains. The cogwheel deagglomerator is operated in rarefied air with a typical pressure of 100 Pa. The single monomer particles couple to the gas on a time scale of $\tau_f \sim 1$ ms and are stopped within a distance of less than 100 mm after leaving the

TABLE I. Properties of the particles used in the experiments.

Physical property	Symbol	Value	Unit	Reference
Material		SiO_2, nonporous		Manufacturer information
Morphology		Spherical		Manufacturer information
Molecular arrangement		Amorphous		Manufacturer information
Density	ρ_0	2.0×10^3	$kg\,m^{-3}$	This work
Radius	s_0	0.76 ± 0.03	μm	[11]
Mass	m_0	$(3.7 \pm 0.4) \times 10^{-15}$	kg	
Surface molecules		Si-OH		Manufacturer information
Surface energy		0.014	$J\,m^{-2}$	[10]
Adhesion force	F_{stick}	$(67 \pm 11) \times 10^{-9}$	N	[10]
Adhesion energy	E_{stick}	$(2.2 \pm 0.4) \times 10^{-15}$	J	[7,9]
Rolling-friction force	F_{roll}	$(0.68 \pm 0.13) \times 10^{-9}$	N	Linear extrapolation from Ref. [10]
Rolling-friction energy	E_{roll}	$(8.1 \pm 1.9) \times 10^{-16}$	J	$E_{roll} = F_{roll} \frac{\pi}{2} s_0$
Sticking threshold velocity	v_{stick}	1.1	$m\,s^{-1}$	Extrapolated from Ref. [7]
Rolling-threshold velocity	v_{roll}	2.2 ± 0.7	$m\,s^{-1}$	$v_{roll} = \sqrt{(10E_{roll})/m_0}$

fast-spinning cogwheel. As the gas is laminarly streaming through the apparatus with a velocity $v_{gas} < v_{hs}$, the monomer grains are slowly transported away from the cogwheel.

Depending on the direction of the streaming gas with respect to the gravitational field (vertically upward or vertically downward), the dust particles reach a gas-permeable substrate with a velocity $v_{im} = v_{gas} \pm v_s < v_{hs}$ where they are deposited in a hit-and-stick behavior. Here, $v_s = g\tau_f$ is the gravitational settling velocity of the monomer particles. Because of the low gas pressure, the particles are in the free molecular flow regime so that no hydrodynamic flow forms in the vicinity of the deposition point. Within a few hours of experiment time, a cylindrical high-porosity dust agglomerate with 25 mm diameter and a thickness of up to $\sim$20 mm is formed [see Fig. 2(a)]. The rarely occurring dust clumps with typical diameters of $\sim$100 μm, stemming from imperfect deagglomeration of the cogwheel, are efficiently prevented from reaching the target agglomerate. In the setup shown in Fig. 1, the gravitational settling of these dust clumps is faster than the upstreaming gas velocity. In an alternative setup, in which the gas flow is vertically downward, an S-shaped syphon serves as an efficient mass filter. The specimen produced by the two alternative setups show no systematic differences in the volume filling factors. The dust samples can be easily transported and manipulated. It turned out that cutting the dust samples by means of a razor blade was achievable without the destruction or partial compaction of the sample [see Fig. 2(b)]. For the determination of the mechanical properties of the agglomerates, we produced cuboidal or cylindrical samples.

The volume filling factor of the samples was determined by measuring volume and mass of each agglomerate. It turned out that the volume filling factor is $\phi = 0.15 \pm 0.01$, in quantitative agreement with the predictions from the numerical simulations in Ref. [5]. Figure 2 shows a typical unprocessed sample (a), a cuboidal speciman (b), a representation of our numerical Monte Carlo simulations of the agglomeration process (c), and a high-resolution SEM image of a sample surface (d).

The close agreement between experimental and theoretical volume filling factors shows that a restructuring of the agglomerate due to gravity-induced rolling of "particle trees," as observed in Ref. [9] is insufficient to considerably change the volume filling factor of the agglomerate. We expect gravitational restructuring of the particle chains when the gravitational torque M_{gr} exceeds the resistance by rolling friction, i.e., if $M_{gr} > s_0 F_{roll}$ (see Table I). The gravitational torque can be expressed by $M_{gr} \approx 2N^2 m_0 g s_0 \sin\alpha$ for linear particle chains containing N spheres and an inclination angle α with respect to the gravitational vector. Thus, we get a maximum chain length that can withstand gravitational restructuring of $l = 2s_0 N = s_0 \sqrt{\frac{2F_{roll}}{m_0 g \sin\alpha}}$. With the particle data from

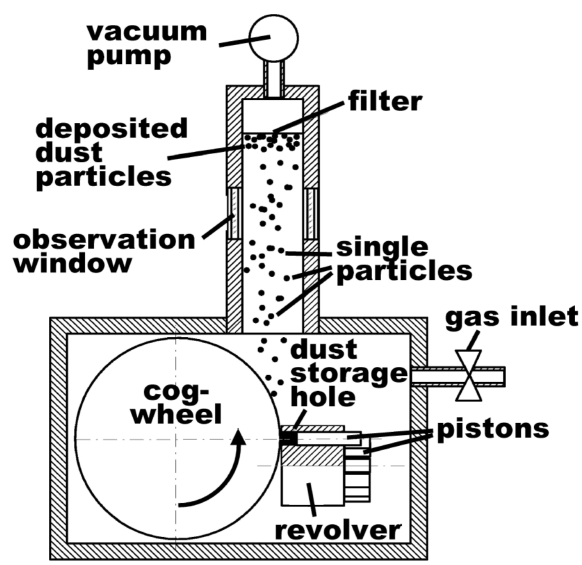

FIG. 1. Schematics of the experimental setup for the formation of macroscopic RBD agglomerates.

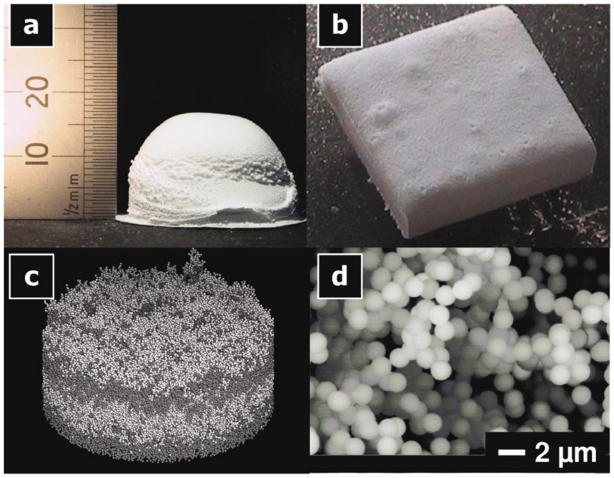

FIG. 2 (color online). (a) An example of an agglomerate with a volume filling factor of $\phi = 0.15$. (b) Specimen of an agglomerate after manual cutting to $\sim 10 \times 10$ mm^2. (c) Result of a Monte Carlo simulation of ballistic deposition. (d) High-resolution scanning electron microscopy (SEM) image of the surface of an agglomerate consisting of SiO_2 spheres with 1.5 μm diameter.

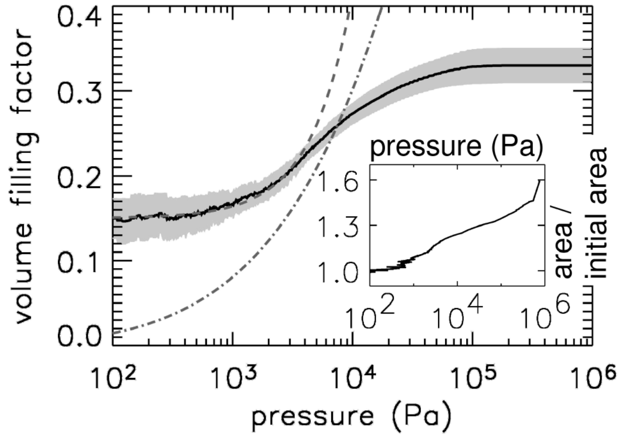

FIG. 3. The volume filling factor of an agglomerate consisting of SiO_2 spheres with 1.5 μm diameter as a function of the unidirectional pressure. The gray-shaded area represents the 1-standard-deviation error of the measurements. The dashed curve is a best-fitting approximation of the form $p \propto (\phi - \phi_u)^\beta$ with $\phi_u = 0.15$ and $\beta = 0.8$, and the dash-dotted curve is the dust-cake compression model by Endo *et al.* [13]. The inset shows an example of the increase in sample cross section with increasing pressure.

Table I and $\sin\alpha \leq 1$, this results in $l \geq 150$ μm, which is in good agreement with measurements of gravitational restructuring of agglomerates in Ref. [9]. The mean free path of a monomer inside an aggregate is $\lambda = \frac{s_0}{3\phi} \approx 1.7$ μm. Thus, a particle chain can tilt only by less than $\sim$1% of its length, so that no considerable change in the volume filling factor results. However, due to the gravitational restructuring, the macroscopic dust agglomerate gets mechanically stabilized.

With the background of understanding the collision behavior of large preplanetary dust agglomerates, we developed an experimental setup for the measurement of the dependence of the volume filling factor on the applied unidirectional pressure. This setup consisted of a quasicylindrical agglomerate sample whose circular faces were connected to a micrometer stage and to a force transducer. Slow motion of the micrometer stage resulted in a gradual compression of the sample. We simultaneously measured the thickness of the compressed agglomerate, its cross-sectional area perpendicular to the direction of the compression, and the applied force. From the latter two, we calculated the applied pressure, and the first two resulted, together with the predetermined mass of the dust sample, in the volume filling factor.

In Fig. 3, the resulting volume filling factor ϕ is plotted as a function of the pressure p. The solid line and the gray-shaded area denote the statistical average of six individual measurements and the standard deviation, respectively. The inset in Fig. 3 shows the increase of the cross-sectional area of one sample as a function of the applied pressure.

As we have seen above, the dust layers have average volume filling factors of $\phi = 0.15 \pm 0.03$ before compression. The larger error for ϕ is due to a lower number of samples used in the compression experiments and due to an indirect measurement of the sample volumes. The data in Fig. 3 shows that below a lower threshold pressure of $\sim$500 Pa, the volume filling factor hardly changes with increasing pressure. Between $\sim$500 Pa and an upper pressure threshold of $\sim 1 \times 10^5$ Pa, the packing density increases steadily from $\phi = 0.15 \pm 0.03$ to $\phi = 0.33 \pm 0.02$. For pressures exceeding $\sim 1 \times 10^5$ Pa, the packing density remains constant at a value of $\phi = 0.33 \pm 0.02$. The relation between compression and porosity given by Endo *et al.* [13] does not fit our data very well (see the dash-dotted curve in Fig. 3). As was previously observed (see Ref. [14] for discussion), the relation between the pressure and the difference between the resulting volume filling factor and its uncompressed value follows a power law $p \propto (\phi - \phi_u)^\beta$ for low pressure values (see Fig. 3), where ϕ_u denotes the volume filling factor of the uncompressed sample. Our best estimate for the exponent is $\beta = 0.8$. Thus, the results are similar to those found by Valverde *et al.* [14] who measured $\beta = 0.824$ for the compression of a sample of toner particles beyond the sample's jamming threshold of $\phi_u = 0.28$.

For application in cometary science, we also determined the tensile strength of the agglomerates. With tensile strength, we denote the minimum force per unit area required to overcome the internal cohesion of the sample. It turned out that the best method for achieving this was to glue two parallel-cut surfaces of a cuboidal agglomerate sample to ultrathin polished glass plates by means of a nonwetting, high-viscosity epoxy resin. The so-prepared samples were introduced into an apparatus similar to the one used in the experiments for the deter-

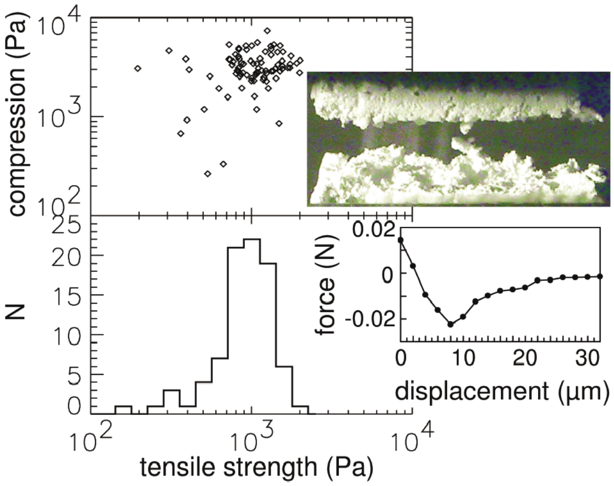

FIG. 4 (color online). Determination of tensile strength for agglomerates. Top: measured tensile strengths of different samples as a function of the compression. Bottom: histogram of measured tensile strengths. The insets show a sample after disruption and a typical force-displacement curve.

mination of compressive strengths. A nonelastic adhesive tape between the outer faces of the glass substrates and the strain-inducing plates of the experiment guaranteed the force transmission to the samples.

For exact parallelization of the samples' surfaces, a compression between ~250 and ~7300 Pa was applied prior to the tensile-strength measurements. Most compressions were in the interval 2000–6000 Pa (see Fig. 4). Because of this initial compression, the samples had average volume filling factors of $\phi \approx 0.2$, slightly higher than their initial values. However, we could not see any strong systematic increase in the tensile strength of the samples with increasing compression (see Fig. 4).

A typical sample after the experimental run is also shown in Fig. 4. In any successful tensile-strength experiment, the samples were disintegrated into two halves of roughly similar thickness, and the separation surface was hill-and-valley shaped. An example of a force-displacement curve of the samples is shown in Fig. 4. With increasing displacement, the initial compression of the sample is relaxed and becomes negative for the strain part of the curve. Within typically 10 μm displacement, the sample reaches the maximum strain and breaks. A logarithmic histogram of the tensile strengths shows a symmetric distribution around a maximum of $Y \approx 1000$ Pa with a FWHM of a factor of 2 (see Fig. 4).

For comparison, we also measured the tensile strengths of highly compressed pellets of 1.3 cm diameter and 0.2–0.5 cm height, which were fabricated in a commercial pharmaceutical pneumatic press with an omnidirectional pressure of $\sim 10^8$ Pa. The volume filling factors of these samples were $\phi = 0.53$–0.55, closer to the RCP. The mean tensile strength of 19 samples was $Y \approx 3700$ Pa. It is interesting to note that the ratio between the tensile strengths of the pellet samples to the agglomerate samples, $\frac{Y_{\text{pellet}}}{Y_{\text{agglomerate}}} \approx 3.7$, is close to the respective ratio of the volume filling factors, $\frac{\phi_{\text{pellet}}}{\phi_{\text{agglomerate}}} \approx 2.6$. Obviously, the number of monomer contacts per unit cross-sectional area determines the tensile strength of a dust agglomerate over a wide range of volume filling factors.

In this Letter we report about a new experiment on the formation of macroscopic agglomerates by random ballistic deposition. Because of the low impact velocities during sample preparation and the large interparticle adhesion forces, the agglomerates formed have the same volume filling factor as in idealized numerical simulations [5] and are stable against gravitational compaction. Compressive strengths of the agglomerates were derived, and it turned out that for low compressions a relation between applied pressure and the volume filling factor in the form $p \propto (\phi - \phi_u)^\beta$ with $\beta \approx 0.8$ exists, in which $\phi_u = 0.15$ is the uncompressed volume filling factor. The tensile strengths of slightly precompressed agglomerates was determined to be $Y = 1000$ Pa.

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*To whom correspondence should be addressed.
Electronic address: j.blum@tu-bs.de

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