Asteroids, Comets, Meteors 1991, pp. 497–500
Lunar and Planetary Institute, Houston, 1992

N93-19228
140970
p. 4

Laboratory Studies on Cometary Crust Formation: The Importance of Sintering

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ABSTRACT

It is demonstrated by experiments and theoretical considerations that sintering processes, so far used to describe the densification of metal and ceramic powders, are relevant for icy materials and therefore probably also for comets. A theoretical model is presented which describes the evolution of so called sinter necks, the contact zone between ice particles. With this model the strength increase of a porous, loosley packed icy body is calculated in which the sinter necks grow by evaporation and condensation of water vapour at a constant temperature. Experiments with ice powders validate the model qualitatively. An increase in strength up to a factor of four is observed during isothermal sintering. In order to check the relevance of the experimental results and the basic theoretical ideas with respect to real comets, more exact theories and improved experiments taking into account additional mass transport mechanisms are needed.

INTRODUCTION

During the experiments within the context of the KOSI (=Kometensimulation)-project two phenomena were observed which are of major importance for the dust emission of comets. First: The near surface observations of ice-/dust-particle emission from the surface of insolated ice/mineral mixtures revealed that the individual particles are bonded together. Before the particles can leave the surface their bonds to neighbours have to be eroded by the gas-jet which is caused from the sublimation of the volatile components, Kochan (1991). Second: The inspection of the material after insolation has shown a remarkable increase in hardness of more than one order of magnitude. Up to now this hardening was attributed solely to the recondensation of the volatiles in colder regions of the sample, Kochan et.al. (1989), leading to a decrease in porosity and an increase in bond area between the particles.

The formation of bonded areas between the ice/mineral particles or agglomerates may, however, not only be caused by recondensation of water vapour at the contact areas but can also occur without insolation by so called sintering processes. Sintering processes of particle agglomerates are a well known and investigated phenomena in the field of powder metallurgy, Ashby(1980). Generally any process leading to a rearrangement of particle agglomerates which is solely driven by curvature dependent gradients of the chemical potential is called sintering.

In this paper we report on experimental and theoretical studies on sintering and show its importance for the comet simulation experiments.

THE PROCESS OF SINTERING

Sintering is driven by the minimization of the free surface energy of an ice particle agglomerate:

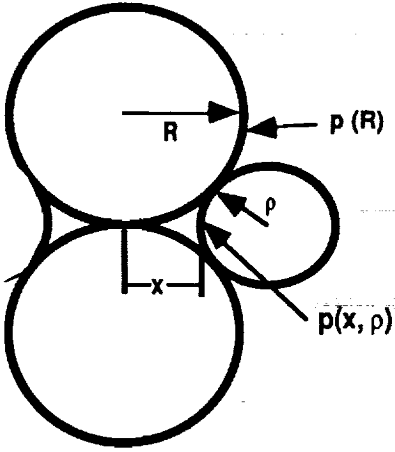
$$G = G(T,p) + \sum \sigma_i A_i \quad \text{with} \quad \delta G = \text{minimum} \tag{1}$$

with G(T,p) the Gibbs free energy of massive ice, T the temperature and p partial pressure. σ_i is the surface tension of the i-th particle and A_i its surface area. The chemical potential of a single particle with radius R which is in thermodynamic equilibrium with its vapour depends linearly according to equation (1) on the particle curvature. Therefore the partial pressure of water above an icy particle depends exponentially on its curvature K:

$$p(R) = p_\infty \exp\left(\frac{\sigma \Omega K}{R_{gas} T}\right) \tag{2}$$

where p_∞ is the partial pressure above a flat surface, Ω the molar volume, K is equal to the sum of the two principal radii of curvature $1/R_1$ and $1/R_2$. R_{gas} is the universal gas constant. Gradients of the partial pressure lead to a transport of vapour within an agglomerate even at isothermal conditions!

If particles touch each other they will develop small neck areas by spontaneous adhesion, Swinkels and Ashby(1980). These necks form zones of concave curvature in contrast to the convex curvature of the spheres. Therefore the partial pressure at the neck areas is smaller than at the remaining areas of the particles (see Fig. 1). The difference in partial pressure leads to a mass transport from the convex to concave surfaces, thereby increasing the neck area. These bridges make the particles sticking together at the surface as well as in the interior.



$$\frac{dV_{neck}}{dt} = z(R,t) \cdot \Omega \cdot A_{neck}$$

$$z(R,t) = \frac{p(\rho,x) - p(R)}{\sqrt{2\pi \cdot \mu_{mol} \cdot R_{gas} T}}$$

z = sublimation rate

A_{neck} = neck surface area

V_{neck} = neck volume

μ_{mol} = molar mass of water vapour

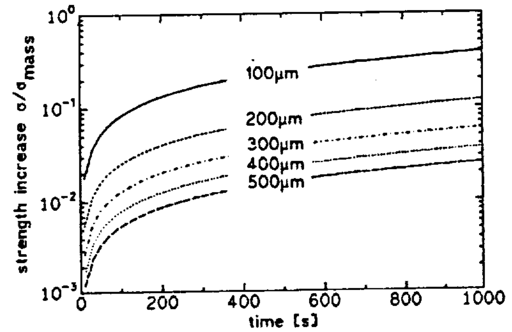
Fig.1: Sinter neck between to particles. p(R) is the partial pressure above the spheres, p(x,ρ) the partial pressure above the concave neck surface.

There are different processes of sintering (see Swinkels and Ashby (1980)). In this paper we confine ourselves to the discussion of the vapour transport mechanism. A calculation of the neck surface area and volume according to the equations shown in Fig.1 reveals, that the neck radius x increases as:

$$x^3 = \frac{3\pi \cdot z_\infty \Omega^2 \sigma \cdot R}{2 \cdot R_{gas} \cdot T} \cdot t \tag{3}$$

if x<<R.Therefore sintering will increase the bonded areas between ice particles in proportion to $t^{2/3}$. The strength of an agglomerate increases proportionally to the

neck area. A calculation of the strength increase of an ice agglomerate due to sintering according to eq.(3) and using the relationship $\sigma=\sigma_{massive}\,(x/R)^2$ for various particle sizes is shown in figure 2 a. Fig.2b shows x(t,R)/R at 240 K without any approximation.The dashed straight lines are the approximation of eq.(3).



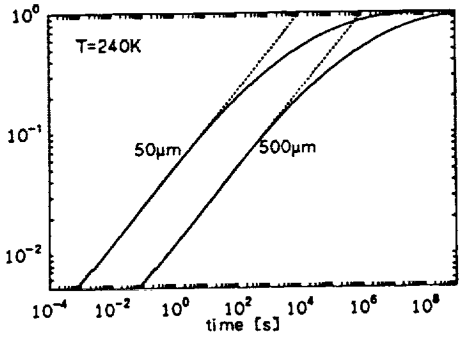
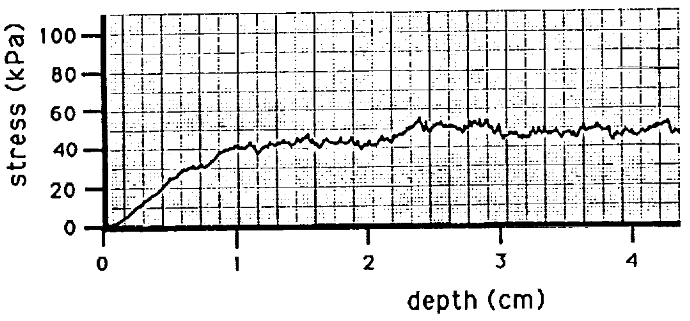


Fig.2a(left): Theoretical relative strength increase of a porous ice body due to sintering.
Fig.2b (right): Sinter neck radius as calculated from an exact theory for different particle sizes.

EXPERIMENTAL RESULTS

In order to validate the considerations made above about sintering of ice particles we performed experiments with porous ice. Spherical ice particles were made by spraying water into liquid nitrogen. The liquid nitrogen then at -20 °C evaporates while the ice powder is continuously stirred to avoid the formation of sinter necks prior to the experiment. The ice powder is filled into copper cans which were thermalized for a period of days to -20°C in a cold lab. The sintering of this isothermal powder as a function of time is examined by the measurement of the strength increase compared to the inititially loosely packed powder bed. Care was taken to ensure the isothermality of the samples and a special procedure to fill the cans was developed making the measurements reproducible. The strength was measured with a spherical indentor which moved into the sample with a speed of 40 mm/min, Kochan et.al. (1989). Fig.3 shows a typical curve of stress versus penetration depth of the indentor. After an initially nearly linear increase in stress a maximum is reached after which the stress oscillates around



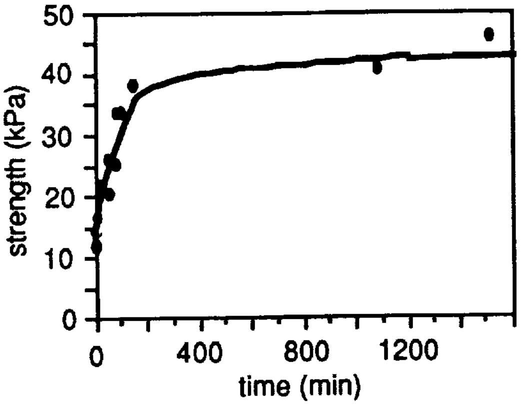


Fig.3(left): Measured stress of a porous ice body as a function of sample depth.
Fig.4(right): Strength of porous ice bodies at -20°C as dependent on sintering time.

a mean value in a random manner. This behaviour is typical for brittel porous bodies (Gibson and Ashby, 1988). The increase of strength as a function of sintering time is shown in fig.4. A comparison of this experimental result with the theoretical prediction of fig.2 qualitatively shows the same behaviour.

Following the two sphere model, used above to calculate the sintering rate as a function of time, we performed experiments with a small number of particles under a microscope. Figures 5 and 6 show the formation of sinter necks between the ice particles directly.

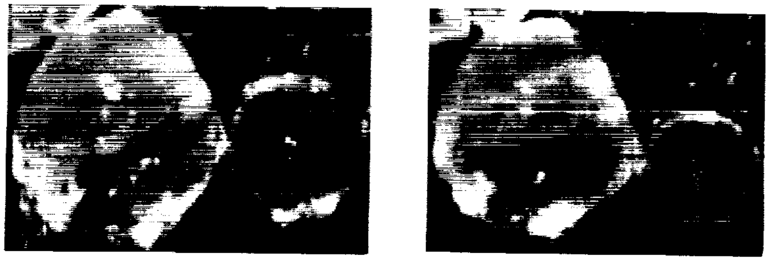


Fig.5 (left): Ice spheres just deposited under a microscope at -20°C. Fig.6 (right): Same arrangement of ice particles after 26h sintering.

CONCLUSIONS

It was demonstrated that sintering processes are even relevant in icy material and therefore most probably also within real comets. The theoretical description given is limited to sinter neck radii which are small compared with the particle radii. Better models are under development, especially taking into account the variation of particle sizes within a sample, which would induce an additional change in the morphology and texture of an ice agglomerate (Ostwald ripening, Voorhees (1985)). In order to check the relevance of these experimental results and the basic theoretical ideas for real comets more exact theories and improved experiments taking into account additional mass transport processes are needed. Further experiments using different porosities and ice-/mineral-mixtures are under preparation.

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