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REPORT

CD NO.

50X1-HUM

COUNTRY USSR

DATE OF
INFORMATION 1950

SUBJECT Scientific - Physics, surface tension

DATE DIST. 24 Oct 1950

HOW
PUBLISHED Monthly periodicalWHERE
PUBLISHED Moscow

NO. OF PAGES 3

DATE
PUBLISHED Jun 1950SUPPLEMENT TO
REPORT NO.

LANGUAGE Russian

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SOURCE Priroda, No 6, 1950, pp 58-60.

ACADEMICIAN LUKIRSKIY'S DISCOVERY
OF IMPORTANT SURFACE TENSION EFFECTS

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In several experiments with acicular monocrystals, it has been noted that the form of the point changes when these ends are heated to quite high temperatures (below the melting point, however). The supposition was that surface tension forces were responsible for this change. Since surface tension of monocrystals depends on crystallographic orientation, we should expect that the monocrystal surface would not return from the pointed to a regular spherical surface.

Academician P. I. Lukirskiy (Doklady Akademii Nauk SSSR, Vol XLVI, 300, 1945) set up a number of experiments to clarify this problem. The experiments were made on quite large monocrystals. Small spheres with diameters ranging from one to several centimeters were cut from rock salt monocrystals. These were then carefully polished, and heated in a furnace for several hours at various temperatures (for different tests) within the interval 720-760° C. Study of the monocrystals after heating established that the monocrystalline spheres took on the form of an irregular hexoctahedron (48 faces) after each such experiment. This effect was obtained after any number of tests with a given sphere; if the polyhedron obtained was again polished and heated, the same figure was reproduced. A slight difference of the lengths of the crystalline axes was observed in the figure obtained from the sphere. For an initial sphere diameter of several centimeters, this difference was only of the order of 10 microns. It was also established that all edges and vertices of the polyhedron obtained were slightly rounded off, so that the transition from one surface area to another was relatively smooth. The phenomenon discovered by P. I. Lukirskiy was explained by the action of surface tension forces.

It was noted above that surface tension in monocrystals depends on the crystallographic orientation selected. Consequently, surface tension of a monocrystalline sphere will not be the same at every point of its surface. It is further known that if atoms of the crystal could move freely, as in the case of a liquid, for example, they would try to occupy a position at which surface energy would be minimum. By keeping the temperature of the crystal

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high, we make possible the freest and most intensive displacement of its atoms, but not to the degree that holds for a liquid. Because of complete isotropy in liquids, the equilibrium figure is a sphere; but other figures represent equilibrium for crystals. Undoubtedly, the hexoctahedron of the rock salt crystal obtained is more "in equilibrium" than a sphere of the same monocrystal. However, as Lukirskiy pointed out, there is still a possibility that the figure obtained is only intermediate and that a final form even more in equilibrium could be obtained. This might occur if there were several solutions to this problem of the free energy minimum, i.e., if several equilibrium forms existed. The last supposition, however, has not been confirmed as yet; despite prolonged heating, no further change in form was obtained.

With regard to the mechanism governing the formation of equilibrium figures, P. I. Lukirskiy noted that this formation might come about through repeated evaporation and condensation or as a result of "creep" of surface atoms. The latter is most probable, since in this case atoms with lower energies could participate in the process.

A very curious phenomena -- straightening out or leveling of the surface -- should take place if creep of atoms is the correct hypothesis. If some face of a crystal is made rough, it should become smooth and shiny after being heated for several hours at temperatures providing intensive creep. P. I. Lukirskiy's experiments showed that this phenomena actually occurred.

The results obtained by P. I. Lukirskiy caused S. V. Starodubtsev and N. I. Timokhina (DAN, Vol LXII, p 619, 1948) to study sintering of monocrystalline powders, where surface tension is assumed to be of great importance. This assumption seems quite plausible; the role of surface tension in monocrystalline powders should be considerably greater than in the case of large monocrystals, since the surface in the first case is much larger for identical volume.

Crystalline powders sinter when the packed powder is heated at a temperature somewhat below the melting point, i.e., the same conditions under which Lukirskiy investigated rock salt monocrystals. Starodubtsev and Timokhina investigated sintering of powders consisting of rock salt crystals (NaCl) and powders containing KI crystals. The powders were prepared in the following way: The salt monocrystals were crushed and then divided into separate fractions with respect to the size of the crystals. The fraction consisting of particles whose linear dimensions varied from 0.1 to 0.075 cm was selected for the experiments. These particles were examined both before and after the experiment in a microscope and were subjected to microphotography when necessary. In the experiment itself, the particles were placed in crucibles and heated for 2 hours at 500° C. The sintered specimen was then again destroyed by application of the proper mechanical load. In the powder thus obtained, there were a great many crystals which had grown together [*narostok*, literally, cluster].

Inspection of the particles which had grown together revealed that they were connected with each other by quite narrow bridges, having a characteristic circular cross section. The width of the bridges was several times less than the dimensions of the particles themselves. This held for particles of NaCl crystals and KI particles. In studying the different connecting bridges, Starodubtsev and Timokhina discovered "embryonic clusters," having the form of thin threads from 300 to 20 microns long, among the couples destroyed by the application of a mechanical load. The smallest diameter of such observable clusters was 20 microns.

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The same authors used another variation of the experiment to study sintering at the very beginning of the process. A small NaCl crystal was placed on one of the planes of a rock salt crystal. The gap between the parallel faces was 20 microns. Both crystals were placed on a quartz plate, which was heated at 600°C. Study of the gap during heating revealed that cone-shaped projections grew from the face of one crystal to the face of the others. Then thread-shaped clusters, which changed into connecting bridges, formed from these cone-shaped projections. It was found that the rate of formation of the clusters was temperature-dependent, and was 3 microns/hr at 600°C for a gap of 20 microns. The experiments also showed that the thread-like clusters frequently do not connect the crystals along the shortest distance. The direction of thread egress depends on crystallographic orientation.

In connection with these experiments, we cite some considerations on the mechanism of the sintering process which were borrowed from a book by Ya. I. Prenkel (Introduction to the Theory of Metals, Gostekhizdat, Moscow/Leningrad, 1948). If all crystals of a powder were instantaneously melted and transformed into drops, all drops would flow together because of the action of surface tension forces. Crystals, just as liquids, although to a considerably lesser degree, have fluidity, provided either by plastic deformation or diffusion flow. We can therefore expect that the particles of a crystalline powder will be drawn together in one mass, a unique conglomerate, at quite high temperatures, when the fluidity of the crystals becomes more noticeable. Of course, considerably more time is required than in the case of liquid drops, since the viscosity of crystals even at high temperatures is considerably greater than the viscosity of liquid drops. If, for example, the viscosity of heated crystals is 10^{10} times that of drops, sintering will be 10^{10} times slower than fusing of drops. Considering the time for fusing of drops to be 10^{-5} seconds, we obtain a rate of growth of the connecting bridges between drops of the order of $5 \cdot 10^5$ microns/sec, or $2 \cdot 10^{10}$ microns/hr for gaps of the order of 50 microns. This figure would yield a rate of growth of the connecting bridges between crystals of the order of 2 microns/hr, which is close to the value obtained in Starodubtsev and Timokhina's experiments.

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