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A TECHNIQUE FOR THE GROWTH OF HIGH QUALITY SINGLE CRYSTALS OF ICE

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Résumé - Une nouvelle technique est décrite pour la fabrication de monocristaux de glace ayant une faible densité de dislocations. Aucun germe cristallin n'est nécessaire et l'eau est gelée de bas en haut de manière à opérer sous vide ou sous atmosphère contrôlée. Des cristaux de 45 mm de diamètre et de 80 mm de long ont ainsi été fabriqués. La densité de dislocations déterminée par topographie X à l'aide du rayonnement synchrotron est inférieure à 100 cm^{-2} .

Abstract - A new technique is described for the growth of single crystals of ice of low dislocation density. No seed crystal is required and the water is frozen from the bottom upwards so that it is possible to work with a vacuum or controlled atmosphere above it. Crystals of 45 mm diameter and 80 mm long have been grown and found by synchrotron radiation topography to have dislocation densities less than about 100 cm^{-2} .

INTRODUCTION

Many workers have developed techniques for the growth of single crystals of ice by the progressive freezing of water. A modified Bridgman method suitable for growing crystals of very low dislocation density has been described by Oguro and Higashi [1,2]. We describe here a technique developed from these methods but including innovations which give it major advantages.

In conventional methods for ice the crystal usually grows above the liquid to take advantage of the natural convectational stability of water. The design has then to permit the expansion associated with the formation of the ice; this normally means that a free liquid surface at atmospheric pressure has to be maintained somewhere. In order easily to be able to grow in vacuum or a controlled atmosphere, we have chosen to grow the ice from the bottom of the container. The system described in [1] and [2] uses a seed crystal, but this is not possible in our arrangement. We have eliminated the need for a seed, and this in fact makes the whole procedure simpler.

APPARATUS

The system used is illustrated in figure 1. The main part of the crystal is grown in the 45 mm diameter section A of the specially shaped pyrex glass container G. This container can be opened at the cone joint C for the removal of the final crystal and is closed at both ends by PTFE valves V_1 and V_2 . It is suspended by a cord in a larger glass tube F, partly filled with an antifreeze solution AF which is kept at a constant level by the overflow OF. The top of the antifreeze is maintained at a constant temperature near to -1°C by the sensor T_1 and heater H_1 . The air in the upper part of F is maintained at a uniform temperature of $+0.5^\circ\text{C}$ by two independent sensors T_2 and T_3 with associated heaters H_2 and H_3 . The whole system is in a larger temperature-controlled enclosure inside a cold room.

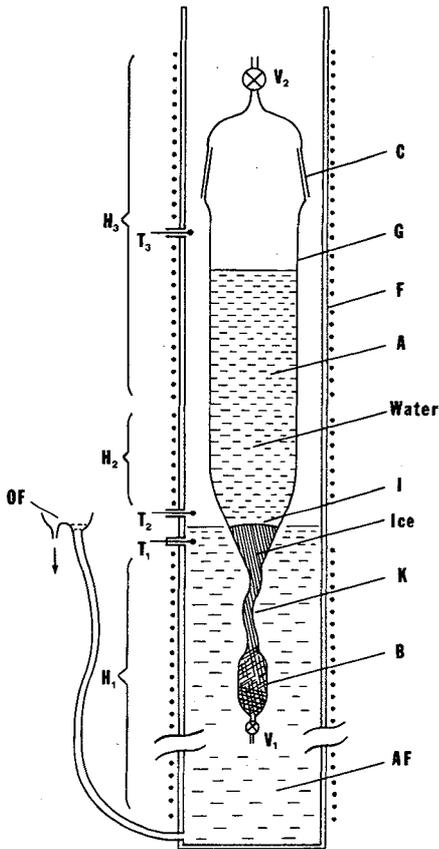


Figure 1. Crystal growth system.
For clarity the diagram
is not drawn to scale.

In operation the growth container G is filled to the level shown with purified and degassed water. It is cooled to near 0°C , and the bottom of the bulb B is dipped into antifreeze at below -10°C until a mass of polycrystalline ice has formed in the lower half of the bulb. Once this has happened the tube is immediately suspended inside the tube F as in figure 1 but with the middle of B at the level of the antifreeze solution. After it has come into thermal equilibrium the growth tube is lowered into the antifreeze at a steady rate of 12 mm per day, and at the same time it is rotated by rotating the suspension cord at a rate of 4 revolutions per day.

As the polycrystalline ice grows up the bulb B a few grains with their *c*-axes approximately horizontal become dominant, and eventually only one of these enters the capillary tube K which contains a kink (as suggested by M. Oguro, private communication). Growth through this capillary eliminates dislocations propagating from B and a crystal of low dislocation density grows up the conical part of G into the region A. When growing in vacuum thorough degassing of the water is important if low pressure bubbles are not to form in the capillary. The temperatures have to be adjusted so that the growth interface I is slightly convex to discourage the growth into the crystal of dislocations formed at the walls. Independent control of the temperatures at T_2 and T_3 is needed to prevent convection currents in the water, which spoil the shape of the interface and may lead to the formation of ice on the top surface of the water.

RESULTS

We have used this technique to grow a number of crystals with a length of 80 mm in the region of 45 mm diameter. Figure 2 is an X-ray topograph taken with synchrotron radiation [3] of a specimen 1.5 mm thick cut from the upper part of a grown crystal. The many dislocations at the bottom of the topograph arise from handling and mounting damage. In the remainder of this specimen the dislocation density is less than about 100 cm^{-2} . Some of the dislocations are in the form of spirals with $[0001]$ Burgers vector [4], which lie in a plane that is perpendicular to the original growth interface. They cannot therefore have been formed during growth, and are thought to have been generated by climb during the thermal history of the crystal.

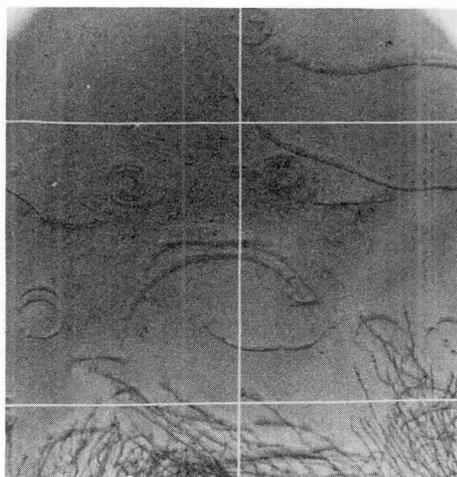


Figure 2. X-ray topograph of specimen cut from upper part of crystal. Plane of projection approximately (0001). Diffraction vector $1\bar{1}00$. Growth direction approximately horizontal in plane of the figure.

Once developed this technique has proved simple to operate, and the density of grown-in dislocations is lower than that introduced by other means. We have yet to assess the chemical purity of the crystals, but the method promises to be as good as anything that can be achieved in glass. Without a seed it is not possible to choose the orientation of the crystal, but this is not essential for successful growth and specimens can subsequently be cut out in whatever orientation is desired.

ACKNOWLEDGEMENTS

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COMMENTS

Question of M.A. WHITE :

How important is it that you rotate the sample as it is cooled, with respect to the defect concentration ?

Answer :

We have not performed experiments specifically to test this. Rotation tends to average out small inhomogeneities in temperature and gives growth which is more axially symmetrical.