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Simple rotating puller for growing single crystals in air and protective atmospheres

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Abstract. A simple equipment for growing single crystals by Czochralski technique is described. The crystals can be grown at any desired rate because of the stepper motor driving a lead screw mechanism. The crystals can also be rotated at a desired rate while pulling. The mechanism employed for this purpose is described. Single crystals of alkali halides measuring 40 mm diameter and 50 mm long were grown using this apparatus. The apparatus can be flushed with Argon in order to grow crystals free from OH^- contamination. Mechanical and infrared transmission properties of these crystals are presented. The adaptation of the apparatus for other types of growth and compounds is discussed.

1. Introduction

Single crystals of alkali halides are commonly used as a starting point for investigations in material research This is because of their ready availability in sufficiently pure form and the ease with which they can be grown. As a result, equipment for growing single crystals of alkali halides are now commercially available since they can be fabricated out of conventional materials. In most of the systems that are commercially available or assembled by individual workers (Buckley 1951 and Brice 1965), the growth rate can not be easily varied and also provision for rotating the crystals while they are pulled is not common.

It is possible to grow single crystals with high perfection, provided flexibility exists in the pulling rate and speed of rotation. This has been demonstrated through the work of Cockayne (1969), Carruthers (1968) and Robertson (1966). There exists a need for the preparation of single crystals of alkali halides in the same system but with parameters that can be varied with each material since their melting points, thermal conductivities and heat capacities differ considerably.

With this in view a simple crystal growing system has been designed using stepper motors for pulling and rotation to provide the desired flexibility. Mechanical properties of crystals grown in such set up are also discussed.

Flexibility in rotation is useful when the crystal growth is made under a slight positive pressure as indicated by Arizumi (1969). Use of dry Ar would reduce OH^- contamination and remove the 2·5 μm band in the infrared when the crystals are used as infrared windows.

2. Experimental

I. *Design details* :

The whole experimental arrangement is shown in the Figure 1. The seed crystal (10) is fixed in the seed holder (9) made of 316 stainless steel which is in turn attached to a tube (4) connected to the lead screw (5). This tube fixed to the nut on the lead screw, can be moved up and down by rotating the lead screw. A rod (3) with holes at some intervals passes through the tube (6) with two longitudinal slots, at 180°. A pin (6) inserted in the slots through one hole of the rod inside enables the tube to rotate with the rod. The rod (3) is coupled to a stepper motor (1) and therefore can be rotated at a desired rate. The lead screw is coupled to another stopper motor (8) through a set of bevel gears (7) and can be rotated at any desired rate. Therefore the tube attached to the nut of the lead screw can be moved up or down at a desired rate.

Using a rod for the seed holder here reduces the growth rate at larger crystal diameters but it also gives a facility of rotating the seed without using water seals which tend to leak and make the crystal growth difficult.

Direct coupling of the motor to the seed holder through a rod facilitates the use of a smaller capacity motor with lesser bulk. Moreover the stepper motors used here offer the advantage of varying the speed of pulling or rotation by just the flick of a switch rather than by changing a few gears, as is the case with A.C. motors. There are some more advantages : (1) the direction of motor can be reversed very easily and (2) the motor does not get heated up even after being operated for long periods. Stepper motors are now commercially available (with 200 steps per resolution, manufactured by M/s Automatic Electric).

The entire assembly discussed is fixed on a rigid frame (15) to reduce vibrations and is kept separate from the furnace (13).

The furnace (13) used is a kanthal wire wound furnace prepared in the laboratory. The temperature is controlled with a proportional temperature controller using SCR. This permits accurate temperature control within ±0·5°C without large fluctuations (upto ±2°C) normally encountered in "ON-OFF" controllers.

II. *Crystal Growth* :

The charge (11) is taken in a ceramic crucible (12) and is kept inside the furnace on a stand ( ). Then the furnace is heated to about 50°C and is left at this temperature overnight to remove moisture from the salt. The next day

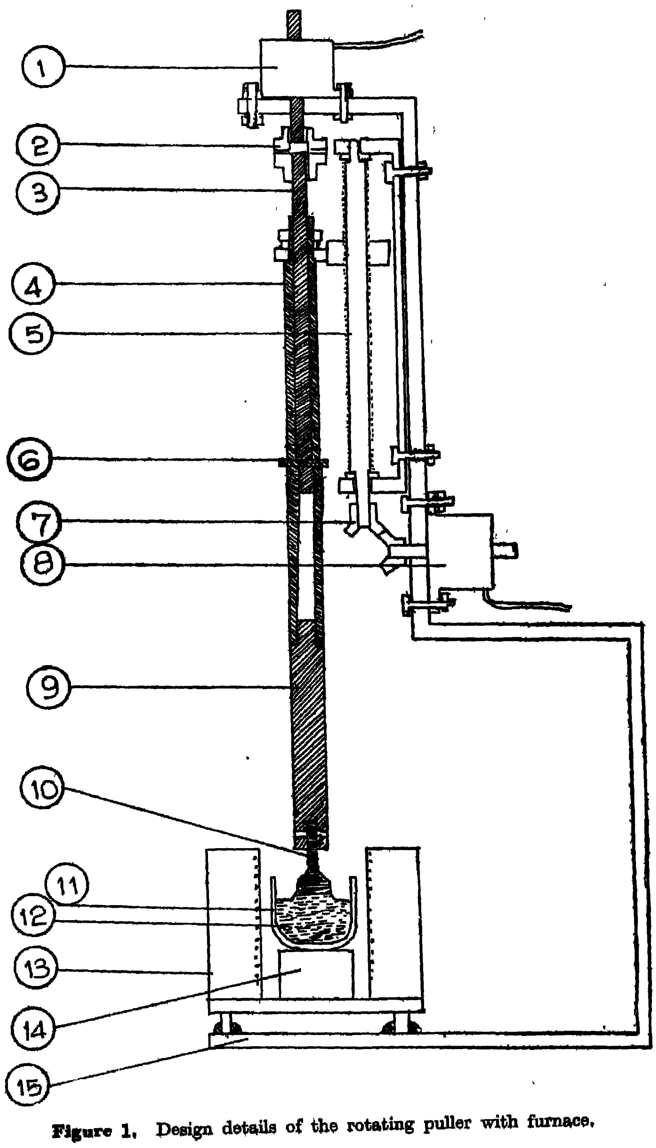

Figure 1. Design details of the rotating puller with furnace.

the temperature of the furnace can be increased at a sufficiently fast rate without risk of hydrolysis of the melt. This is particularly good for a humid place like Bombay.

The crystal seed (10) is fixed in the crystal holder (9) and is slowly lowered inside the furnace to avoid thermal shocks. When the melt aquires a steady temperature a few degrees ($\approx$ 5–6°C) above the melting point, the seed is lowered at a faster rate first and at a very slow rate (3 cm/hr) after it reaches the surfaces of the melt. As soon as the seed makes a splash in the melt, the pulling motor is kept to the reverse mode and the seed is taken up by about 4 mm from the point of the contact observed. The point of contact can be observed by the reflection when light is focussed on to the solid liquid interface, from which is seen a miniscus. Now the lifting is stopped for a while by putting off the pulling motor, and rotation of the rod is started at a speed of 20 r.p.m. When the meniscus spreads a upto a few mm from the seed, the lifting up of the seed is again started at a speed of 1·5 cm/hr. The diameter of the growing crystal can be controlled by adjusting the temperature efficiently. If the temperature is higher than required, the size of the growing crystal tends to reduce resulting in a tapered crystal boule and finally breaking contact with the melt. If the temperature is too low, spurious nucleation starts at the surface of the melt providing many nuclei and resulting in polycrystalline boul. Therefore just the right temperature only gives the best single crystal. It is possible to grow highly uniform crystals by keeping a reference point to view the reflection from the miniscus.

In most of the growth runs conducted with NaCl and KCl, the crystals were found to exhibit morphological facets. The crystals do not grow as cylinders but as rectangular slabs.

III. *Results* :

Figure 2 shows the photograph of a NaCl crystal grown in this set up. It shows the changes in the section of the crystal caused by changes in the temperature. A crystal plate cleaved along an axis perpendicular to the growth axis is shown alongside (Figure 2). This shows the perfection of the growth when a stepper motor is used for rotation.

Dislocation etch pits produced on the cleaved faces exhibit a density of the order of 10^5 dislocations/cm^2. The annealing at 400°C has been observed to reduce the dislocation density while the introduction of dopants in small amounts (0·01 to 0·1% by weight) caused an increase upto 10 times resulting in the dislocation density of the order of 10^6 dislocations/cm^2. Effect of some divalent ions on the hardness has been studied as the divalent impurities are known to harden alkali halide crystals.

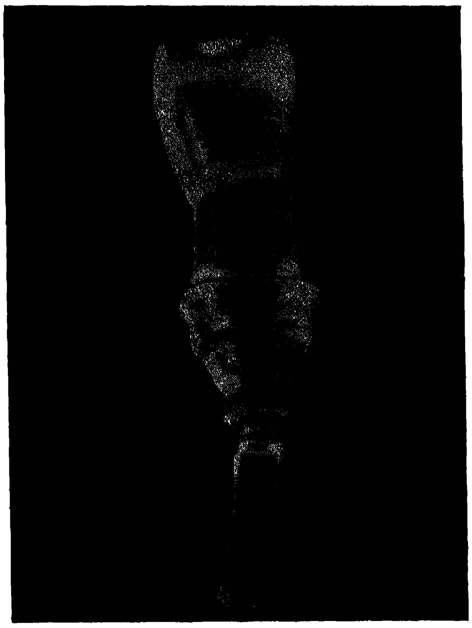

Figure 2. Single crystal of NaCl growth in the set up.

Figure 3. Dislocation etch pits on the (100) faces of NaCl crystals grown in the set up. (a) As grown, (b) annealed at 400°C.

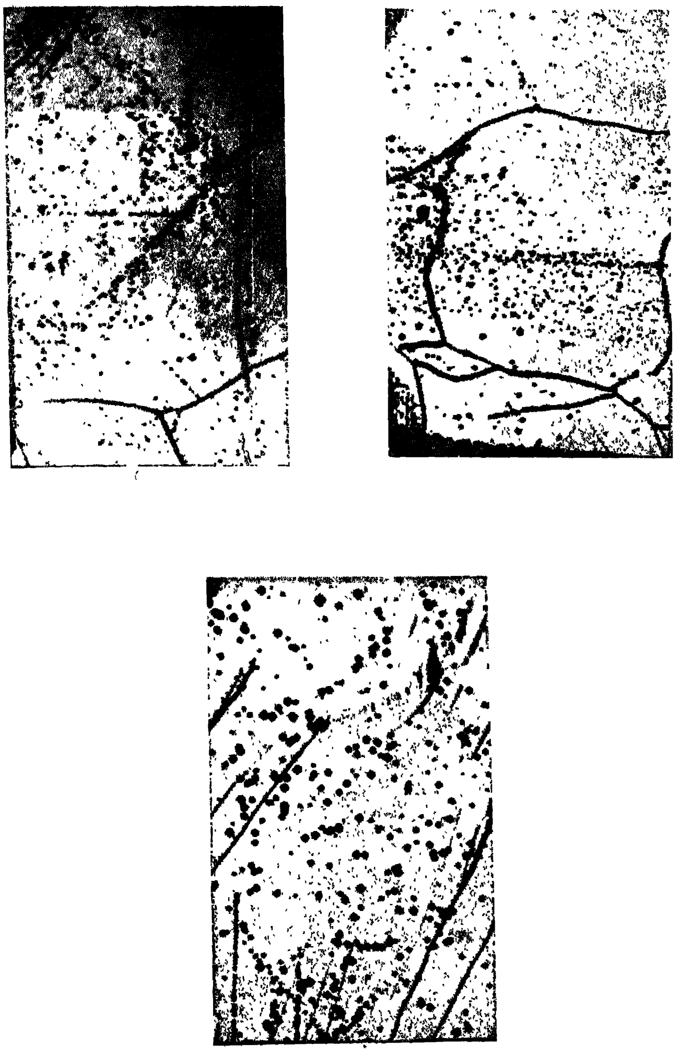

Figure 4. Variation of etch pit density and distribution with dopants (a) NaCl : Mo, (b) Ca, [illegible]

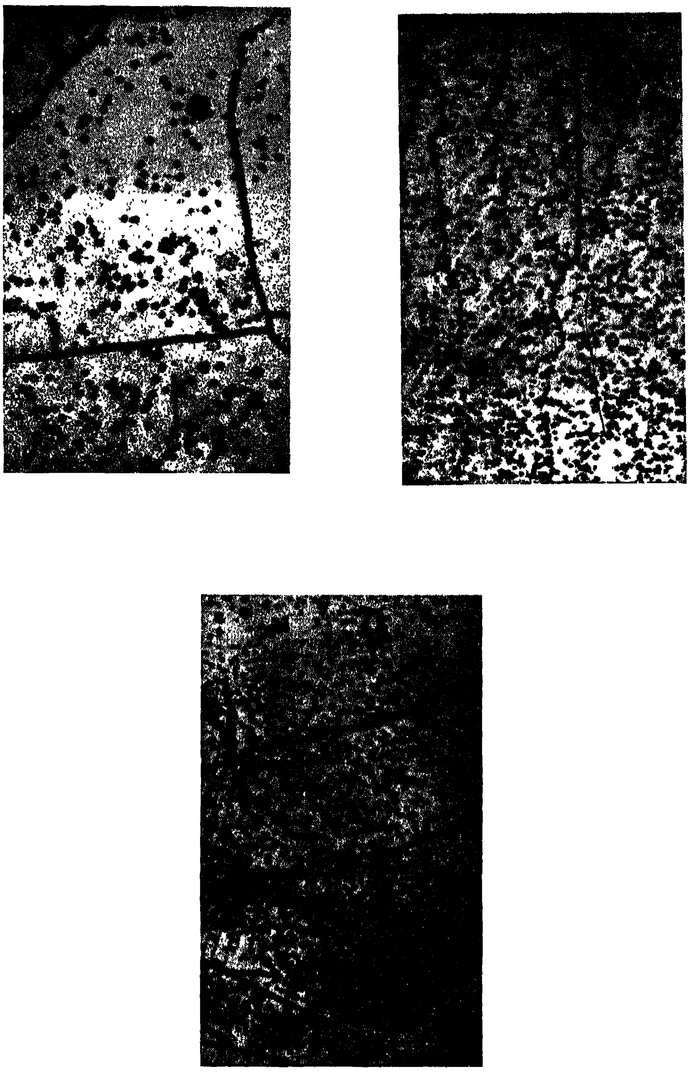

Figure 5. Variation of etch pit density with Ca concentration. (a) 0·01 Wt% [10^4 Pits/cm^2), (b) 0·05 [10^5] (c) 0·1 [10^5 to 10^6].

The effects of varying the dopant and the concentration of a particular dopant on the microhardness of the crystal are depited in tables 1 and 2.

The addition of Ca^{++} caused the maximum hardening, Mg^{++} came the next and Mo^{++} did not change the hardness appreciably. The hardness was found to increase with the increase in Ca^{++} concentration. These results are pertinent in the design of windows for lasers as the increase in the hardness would permit the use of thinner windows reducing the absorption, provided the dopant does not introduce unwanted imperfections.

Table 1.

Crystal	Dopant	Concentration in % by weight	Vickers Hardness, Hv in Kg/mm²
NaCl	Ca	·05	21·30
NaCl	Mg	·05	19·37
NaCl	Mo	(·01)	16·99

Table 2.

Crystal	Dopant	Concentration in % by weight	Vickers Hardness Hv in Kg/mm²
NaCl	Ca	·016	18·15
NaCl	Ca	·05	21·30
NaCl	Ca	·1	22·53

Plates of NaCl and KCl cleaved out of the single crystals grown in this set up did not exhibit any absorption due to OH^- at 2·7 μm. This indicates that the procedures described in the present work could result in good crystals useful for most spectroscopic application. Their usefulness in as Laser windows can be assessed only through Laser Calorimatry which is being developed in the laboratory presently.

Preparation of Crystals under protective atmospheres :

This system can be adapted to grow Crystals under a slight positive pressure of a protective atmosphere also. A quartz envelope with a Wilson seal is placed into the furnace to enclose the crucible. The pulling rod is introduced into the system through the Wilson seal. The process can be repeated to grow crystals as described earlier.

Acknowledgments

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