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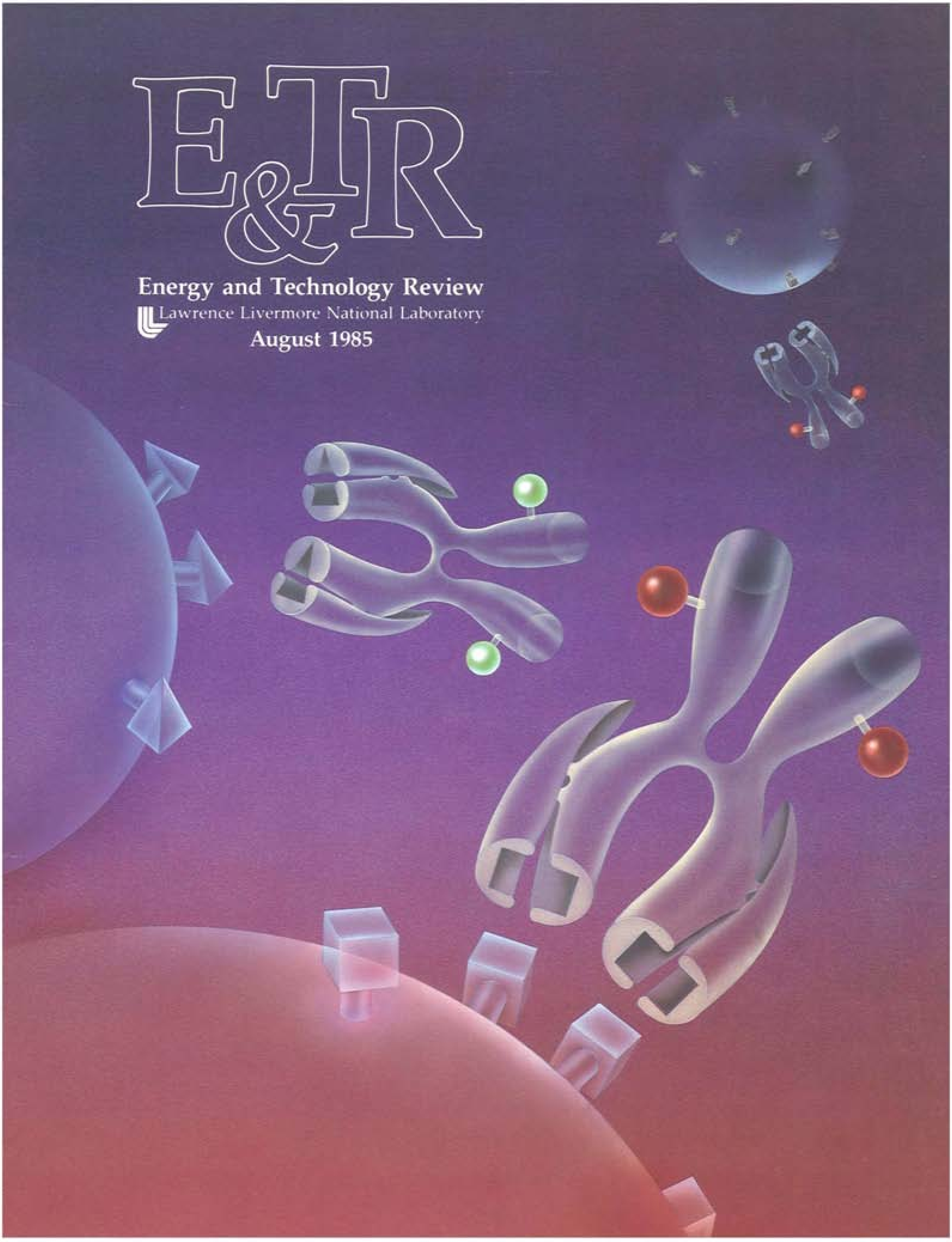

Rapid Growth of KDP Crystals

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In a program aimed at reducing the cost of a possible next-generation fusion-research laser driver, we are developing a new, fast process for growing the large potassium dihydrogen phosphate (KDP) crystals used for optical switching and frequency conversion on high-power laser systems like Nova. This new process grows the crystals ten times faster than the current industrial process at greatly reduced cost. We also seek to produce crystals that are more resistant to damage from intense laser light.

A considerable portion of the cost of high-power neodymium-glass lasers for fusion research is for optical crystals. For example, the KDP crystals used in the Nova laser cost about $20 per cubic centimetre, and tens of thousands of cubic centimetres are used. Such large volumes are needed because the KDP components must have both a large areal extent (to accommodate the large-diameter laser beams) and a considerable thickness for the beams to pass through (to give high conversion efficiency). The large beam diameter is necessitated by KDP's low resistance to damage by passage of the laser light; to avoid damaging the KDP, the beam intensity per unit area must be kept relatively low, which means that the beam diameter must be large.

Two approaches are apparent for cutting the cost of the KDP crystals (they are actually interrelated, but we consider them separately in the interest of clarity). One approach is to cut the cost of producing the crystals, which can be done most readily by increasing the growth rate of the material from 1.5 to 15 mm/day. To date, most of our effort has been spent on this approach. The other approach is to increase the crystal's damage resistance (currently about 7 J/cm^2), making it possible to use smaller-diameter laser beams and, hence, smaller arrays of KDP crystals. We have done some initial investigations on this second approach.

We have been studying the growth of optical crystals in general and KDP crystals in particular. Our immediate objective is to find ways to reduce the cost of the KDP elements by about an order of magnitude. Our broader objective is to advance the scientific and engineering basis for economical growth of optical crystals, whether from supersaturated solutions, as with KDP, or from high-temperature fluxes, which have similar growth-rate limitations.

KDP crystals are grown industrially from large seed blocks, 0.3 m on a side, placed in tanks containing saturated solutions of KDP in water. The temperature is gradually lowered to create a supersaturated solution, and the excess of KDP above the equilibrium level provides the driving force for crystal growth. At the current industrial growth rate of 1.5 mm/day, it takes nearly a year to grow a large boule of KDP (Fig. 1). Increasing the growth rate tenfold would decrease the production cost by a similar factor, assuming that the cost of the required hardware remains unchanged.

We analyzed the industrial process and practice of growing KDP crystals and identified three major barriers to fast growth: (1) impeded transport of dissolved KDP from the bulk of the growing solution to the crystal growth surfaces, (2) insufficient supersaturation of the growing solution, and (3) inaccurate measurement of supersaturation, leading to failure to control it. We have addressed these problems in our research, and the solutions we obtained have enabled us to develop a much faster process for growing KDP crystals of comparable quality.

In our new process, we have improved the transport of dissolved KDP

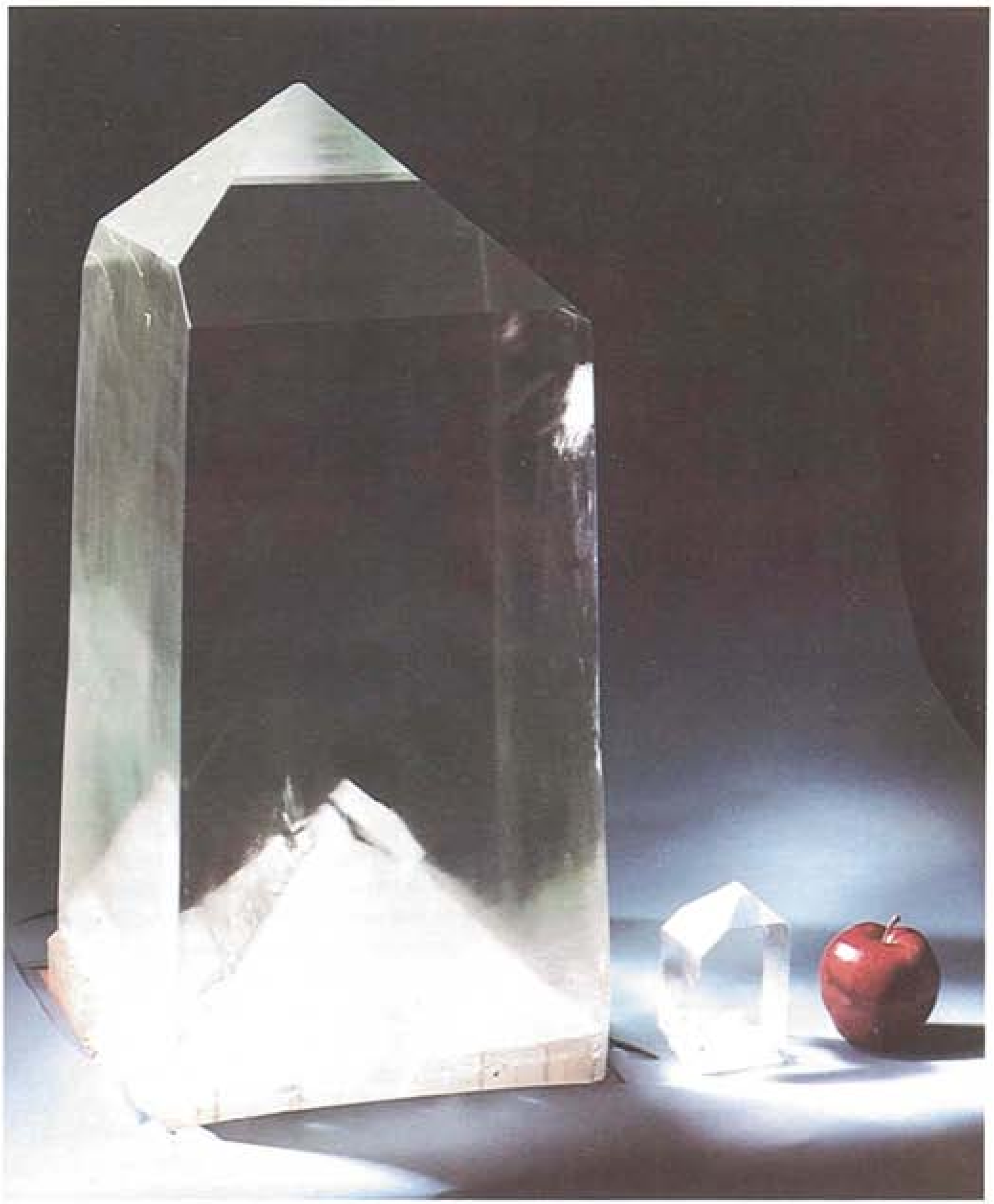

Fig. 1

The boule of KDP (at left), grown by Cleveland Crystals, is large enough to yield a 27- by 27-cm crystal of the size required for the Nova frequency conversion arrays. It took nearly a year to grow this large KDP crystal using the current industrial process.

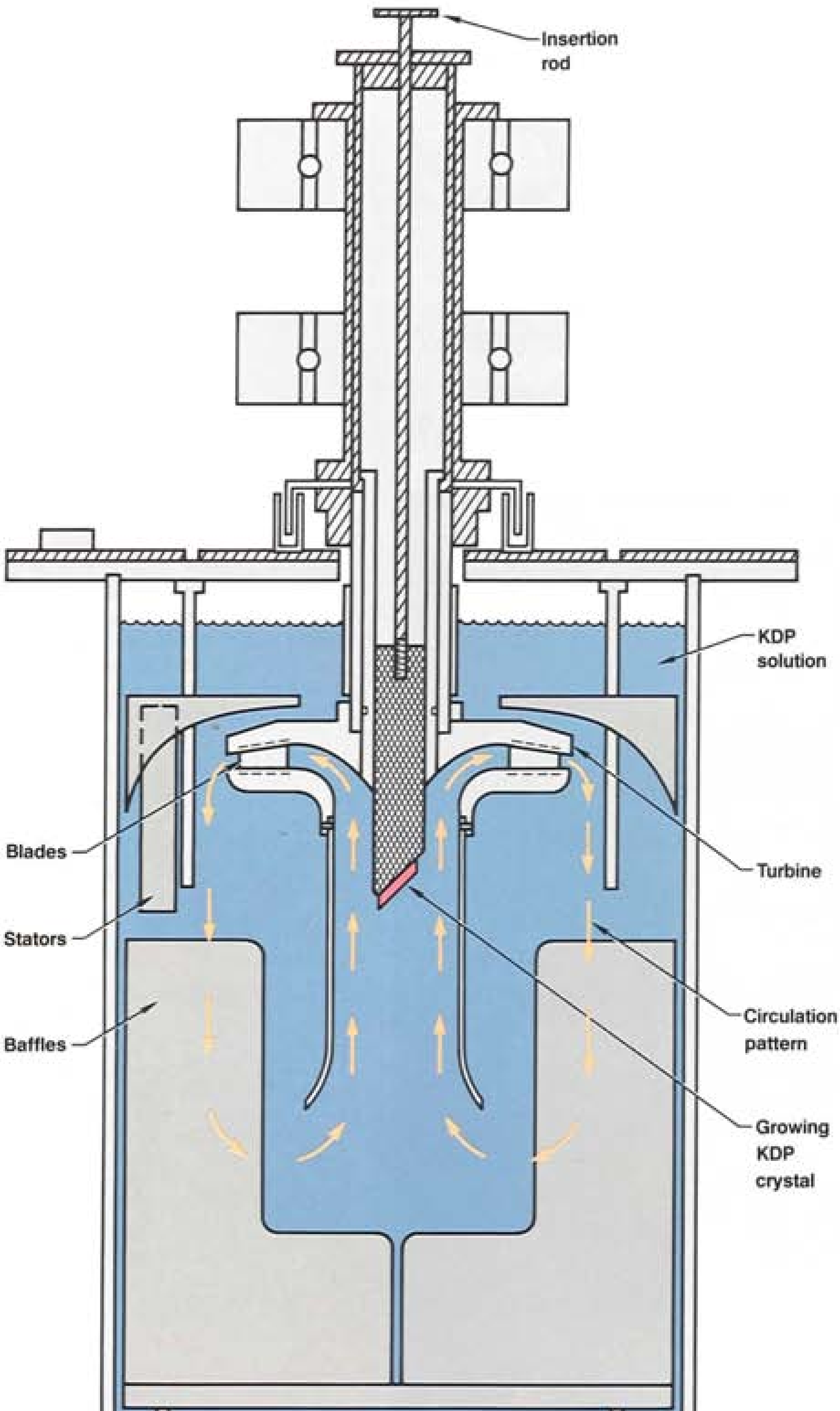

Fig. 2

Turbine apparatus for growing KDP crystals at an accelerated rate. The slowly turning turbine sets up a circulation pattern in the supersaturated KDP solution, providing a constant supply of fresh KDP solution to the crystal growth surfaces. The tank capacity is 21 litres; the growing crystal face dimension is 20 mm.

to the crystal growth surface by placing the seed crystal in the center of a slowly rotating turbine in the solution tank (Fig. 2). The circulation pattern established by the turbine blades draws the solution up into the vertical tube of the turbine, where it flows freely against the (101) crystal growth faces, increasing the flux of KDP across the diffusion boundary layer and thus greatly speeding the crystal growth. The turbine is shown in Fig. 3a. With this turbine circulation system, we have grown KDP crystals at rates up to 25 mm/day without degradation of optical quality.

We have developed a mass-transport kinetic model, based on published growth-rate data on 3-mm crystals and on hydrodynamic considerations, that enables us to analyze the crystal growth process. With this model, we can calculate for various flow configurations the temperature and concentration required for specific growth rates. The growth rates and net growth predicted by our model are in good agreement with the actual growth rate of a 20-mm crystal (20 mm/day using the turbine circulation system).

Our rapid-growth process scales according to the ratio of the solution flow rate to the crystal dimension; larger crystals require higher flow rates. We calculate that a large, Nova-scale crystal would require a flow rate of at least 0.6 m/s.

The turbine shown in Fig. 3b is designed for greater hydraulic efficiency, an important factor in suppressing the undesirable tendency toward spontaneous nucleation. Spontaneous nucleation of minute crystals causes a multitude of problems. These tiny crystals block the tubes in the turbine and flow system, their formation robs the growing crystal of material, and the tiny crystals can stick to the surface of the growing crystal and degrade its optical quality.

This turbine, made of inert polysulfone and Teflon, provides for orienting the crystal with respect to the solution flow, thus allowing control over the origin and direction of the crystal growth steps; it can handle crystals up to 40 mm on a side. We are using this second turbine to investigate the influence of hydrodynamics, growth mechanisms, and impurities during the crystal growth process on the susceptibility of the finished crystal to damage by laser light.

In our efforts to grow crystals that are more resistant to laser damage, we are examining the effects of various growth conditions. For example, we have varied the growth rate from 5 to 30 mm/day, the solution temperature from 40 to 70°C, and the solution pH. We have experimented with controlling growth morphology and the movement of growth macrosteps in order to prevent collision of the growth steps (which causes a defect in the crystal) and subsequent entrapment of impurities. We are working to reduce the size (thickness) of growth macrosteps by isolating impurities that cause their formation. We may be able to use complexing agents such as EDTA, which isolates transition metal ions by forming stable complexes with them, to prevent the adsorption of impurities onto the crystal growth surface. We also are looking into the effects that defect concentrations in the seed crystal may have on the growth morphology.

We are constructing a pilot plant at LLNL for demonstrating our rapid-growth process at one-third scale. The turbine to be used in the pilot plant has an external housing to efficiently convert centrifugal fluid flow into a jet that is directed downward onto a stationary seed block 0.1 m on an edge.

To deal with the other problems we found with the current industrial process of growing KDP crystals (e.g., inadequate measurement and control of supersaturation), we have designed an advanced growth system with separate chambers for crystal growth, preparation of the saturated KDP solution, and elimination of nuclei. We have determined the heat and mass flows required to sustain supersaturation in this system. We have also developed techniques for *in situ* measurement of supersaturation, through differential measurements of solution buoyancy, density, or conductivity, that will allow better control of the level of supersaturation.

If we can successfully translate these features into a practical industrial process of low capital cost, we should meet our immediate objective of cutting the production cost of KDP crystals by an order of magnitude.

Key Words: crystal growth; laser damage; potassium dihydrogen phosphate (KDP).

Fig. 3

Turbines used to grow KDP crystals at rates ten times faster than in the current industrial process. The turbine in (a) was used to provide a free flow of the supersaturated KDP solution to the crystal growth surface; a growth rate greater than 20 mm/day was achieved with this turbine. The turbine in (b) was designed for greater hydraulic efficiency and allows for control of the origin and direction of growth steps, factors that may be important in growing damage-resistance crystals; we are using this turbine in growth studies on larger crystals.