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Contributed Paper

Growth of Bulk KDP and DKDP Crystals

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ABSTRACT

Bulk KDP crystals(45×45cm^2×60cm) were grown by solution circulating method. The relationship between optical homogeneity and growth conditions have been investigated. The way of increasing laser damage threshold was found. Large Type I,Type II and switch have been fabricated using these crystals.

Keywords: Bulk KDP & DKDP crystal, ICF, solution circulating method.

1. INTRODUCTION

Inertial confinement fusion (ICF) engineering need large plates of NLO crystals for electro-optic switches and frequency conversion . Usually ICF requires a single crystal with linear dimensions in the 50cm range. KDP(KH_2PO_4) and DKDP($K(D_xH_{1-x})_2PO_4$) are the only materials used for ICF due to their exclusive physical properties. These crystals have wide transparency region(0.2-1.2μm) and resistance to damage by laser radiation , and relatively high nonlinear efficiency, They are also easily grown into single crystals having large size and high optical quality.

The growth method and growth conditions of high optical quality crystals as well as the relationship between laser damage threshold and optical homogeneity have been investigated in this report.

2. CRYSTAL GROWTH

2.1 Traditional growth method

The lowering temperature growth from KDP solution, growth rate is low, 0.5-1mm/day. Slow growth leads to growth crystal period exceeding one year. It is very difficult to get large size(50×50×50cm) crystals.

2.2 Solution circulating method

It is very difficult to grow bulk crystals by traditional lowering temperature method. We used four tanks (growth tank, Saturation tank, Superheating tank, Buffer tank) to grow bulk crystals by solution circulating method. The stability of growth solution is high, so spoutaneous nucleation and some defects formation could be avoided. The growth rate was about 2-5 mm/day.

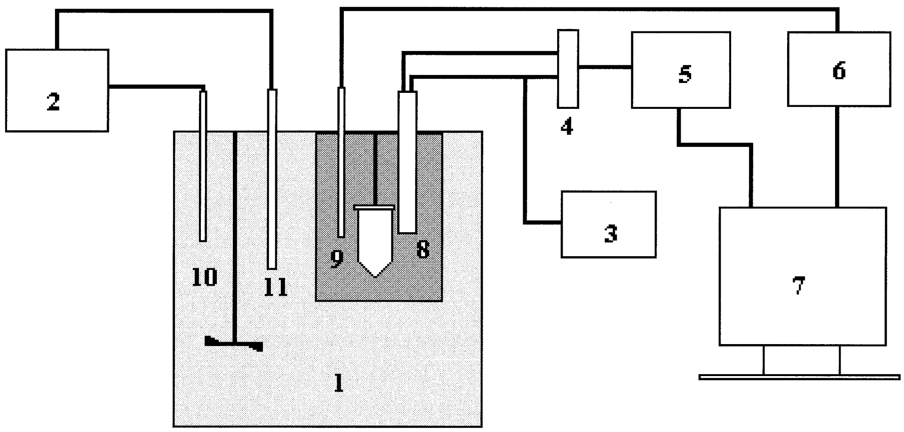
Real-time cross-correlation between concentration, temperature and electrical conductivity of KDP aqueous solution were determined by using transformer-type conductivity sensor. The correlation of solution saturation temperature Ts and conductivity L

$$L(Ts\bullet t)=L_0(t)+b_1(t)+b_2(t)Ts_2$$

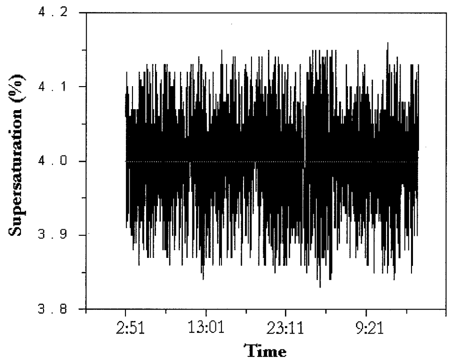
On the basis of this correlation, the computer was made to realize the in-time measurement and control of the supersaturation of the growth solution. This system can accurately control the supersaturator, as shown in Figure 1.

3. MAIN DEFECTS

Scatter particles in KDP and DKDP crystals were measured by ultra-microscopy, TEM and AFM. It can cause light scattering and lower the damage threshold of crystals.



(a) block diagram of the setup for conductivity measurement 1. crystallizer; 2,6.Programmable controller; 3.Digital function generator 4.Scan 5.Digital multimeter; 7.Computer; 8.Conductivity probe 9,10. Temperature probe; 11.Heater.



(b) supersaturation stability obatained by using computer control system.

Figure 1. KDP crystal growth by lowering temperature using supersaturation-control system.

(1) Scatter particles:

The light scattering in crystals was detected by laser. The laser damage threshold of crystal is related to the density of scatter centers; the higher the density of scatter particles, the lower the damage threshold, Usually, the scatter particles size is 10-100 nm.

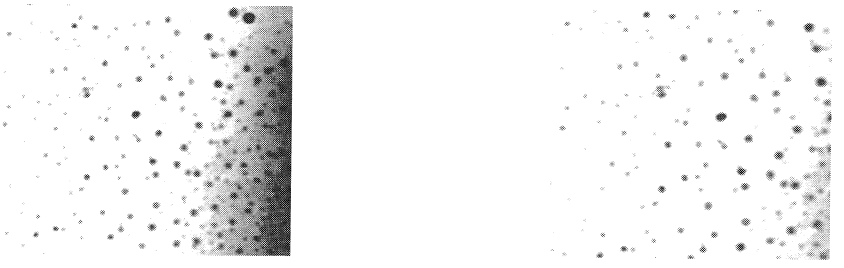
The scatter particles were distinguished into point scatterers, and line scatterers, rile scatterers according to the shape and size of scatter particles. Three kinds scatter particles were observed by TEM.

(2) The scatter particles of crystals grown from different raw materials were shown in Figure 2

(3) There are some impurities existing in the solution. They have various effects on the habit of growth and scatterers. The effects of cationic ions, such Fe^{3+},Cr^{3+},Al^{3+} etc. have been extensively investigated. Reports on the effects of anionic ions are rarely seen but there are some indications that some anionic ions such as H_3BO_3, $H_4P_2O_7$ etc can easily cause scatter particles to form in crystals. The results shown in Figure 3

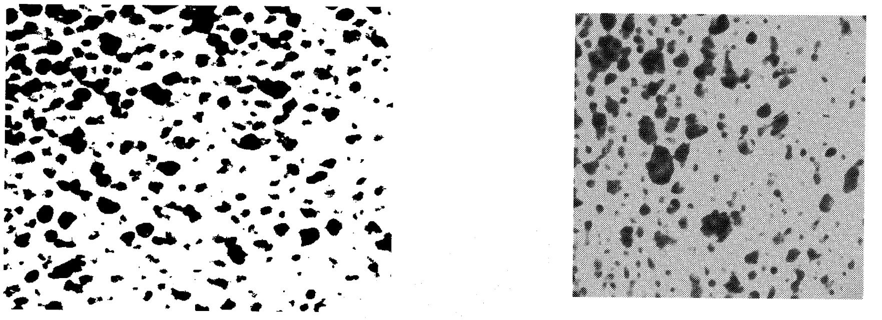
(4) The effect of pH value on the scatter particles:

The pH of the solution have some effects on the scattering behavior of crystals. Usually, single crystals were grown from PH 3-4



(a) analytic grade (b) special pure grade

Figure 2. TEM pictures of scatter particles, the number of scatterers of Condition of (a) is more than that of condition (b).



(a) doped H3BO3 TEM result (b) doped H2P4O7 TEM result

Figure 3. Anion ion group formed scatter particles.

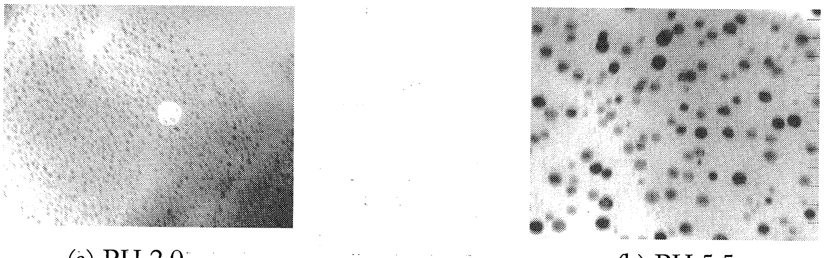
solution. The crystal prism sector will be extended when PH value is too low(2.0) or too high(5.5). The scatter particles will be increased. The results of TEM were shown in Figure 4.

(5)The effect of supersaturation on scatter particles:

Usually, the scatter particles of crystals grown in the same solution but at higher super saturation will have higher density. The scatter particles were obviously increased if the growth rate was over 10 mm/day (in ordinary solution). The results of observation were shown in Figure 5.

(6)Impurities and scatter particles:

The impurities in the solution were an important source which caused scatter particles to form in grown crystals. Some impurities were absorbed on the growth steps to form inclusions (scatter particles). Growth steps were formed on the growth surface as shown in an AFM image(Figure 6).



(a) PH 2.0 (b) PH 5.5

Figure 4. The effect of pH value on the scatter particles.

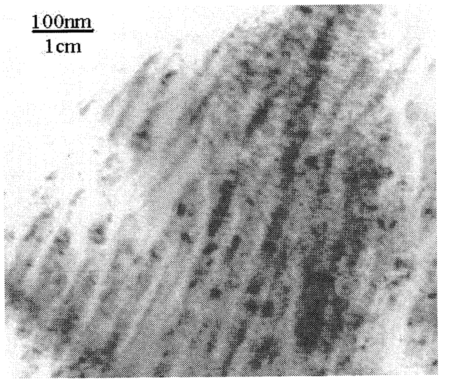


Figure 5. The effect of super saturation on scatter particles.

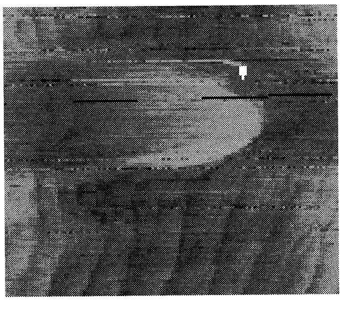


Figure 6. AFM results of formed step.

4. The Methods of Decresing Scatter Particles

In order to growth high quality crystals, the following growth conditions are required.

(1) using high-purity raw materials and optimum growth conditions.

(2) the cycle filtration solution

(3) annealing of crystal etc.

5. Growth of Different Deuterium Content DKDP

(1) high deuterium DKDP(~98%)

Synthesis of DKDP materials:

$P_2O_5+D_2O \rightarrow D_3PO_4$

$D_3PO_4 + K_2CO_3 \rightarrow KD_2PO_4$(>98%)

(2) Different deuterium content DKDP crystals-dissoluble KDP

$KH_2PO_4+D_2O \rightarrow K(H_xD_{1-x})_2PO_4$

(3) Growth of DKDP crystal

Growth method-lowering temperature method

6. Conclusions

Bulk KDP and DKDP crystals have been grown in our Lab by solution circulating method. Crystal size of 45×45×50-60cm^3 could be obtained. The properties were shown in Table 1. The grown crystals and their transparency measurements were shown in

Table 1. The properties of crystals.

	transparency	loss	Damage threshold	Δn
KDP	90%(1.05μ)	<5%/cm	>15J/cm^2	$<10^{-5}$
DKDP	91%(1.05μ)	<1%/cm	>10J/cm^2	$<10^{-5}$

Figure 7 and Figure 8. These crystals have good optical quality and were used to fabricate Type I,Type II and switcher devices.

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Figure 7. bulk KDP crystal

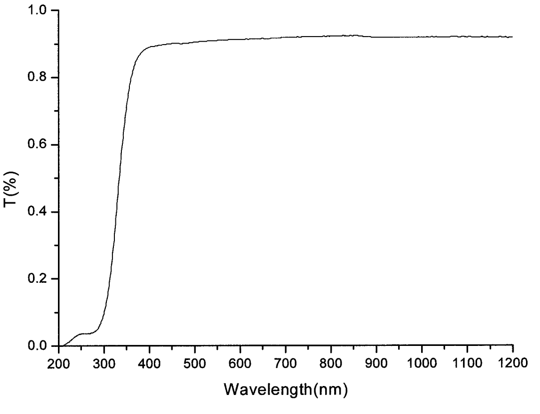

Figure 8. The transparency of KDP

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