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Growth of Technologically Significantbulk Crystalsby Unidirectional S-R Method: A Review

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Abstract : The Review of Literature plays a vital role in all Researches. It is the initial work for carrying out a focused innovative research in a specific field. It empowers a Research intellectual to become an expert in his/herresearch area. It also provides a basis of deep knowledge in research field. It will give summary of available literature of particular area of research. Sankaranarayanan-Ramasamy (SR) method was discovered recently which comes under low temperature solution growth of single crystals. Since then, various developments were made on SR method and lot of researchers are doing research in that method still today in various laboratories throughout the globe. It is a novel method to grow large size crystals. By this method, single crystals shall be grown in a particular orientation. Simple and less expensive experimental set up of this method can be easily installed in any laboratory. Positive, Negative, High, Moderate and Low solubility materials shall be grown by SR method. Modified SR Method, Vacuum Assisted SR Method are the different types of SR Method. The Rotational SR Method (RSR) is the latest modification in SR Method. In this Review, Growth of Technologically Significant Bulk Crystals by Unidirectional S-R Methods were reviewed.

Key Words : Review, Bulk Crystals, Unidirectional Growth, S-R method, Technological Significance.

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1. Introduction

Uniform spatial arrangement of atoms in a matter constitutes crystal. If every atom is surrounded by a precisely uniform spatial arrangement of other atoms and free from inner boundaries, then it is called as an ideal crystal [1]. Crystal growth is the process by which a crystal grows through continuous addition of atoms or molecules to the surface of a seed crystal. The crystal growth process involves a transition of three states namely solid-solid, liquid-solid and vapour-solid [2]. In each system, the crystallization can take place through a formation of new phase with three dimensional nuclei. The most important phenomenon in crystal growth is nucleation [3].Since, crystal growth is a non-equilibrium process, attention must be given to the parameters like concentration, solubility, temperature and other gradients [4].

2. Crystal Growth from Solution

Crystal growth is a challenging task and the method followed for crystal growth depends upon the characteristics of the materials such as its melting point, volatility, solubility and so on. Growth from solution is more widely used to grow the crystals, which have high solubility and have variation in solubility with temperature[5].The method of crystal growth from low temperature aqueous solutions is extremely popular in the production of many technologically important crystals. Crystal growth plays an important role in the area of medicine, engineering, technology and also in the areas of defense and space science. Due to the developments in the field of crystal growth and awareness about structural properties which related to molecular level, there is a great advancement in science and technology [6].

3. Sankaranarayanan -Ramasamy (S-R) Crystal Growth Method

Sankaranarayanan and Ramasamy method has been developed to grow large size single crystal in a particular direction [7].It is one of the methods to grow the crystals from solution. In Sankaranarayanan-Ramasamy Method, the experimental set up is made up of a glass ampoule with a tapered V-shaped or flat bottom portion to mount the seed crystal and a U-shaped top portion to fill saturated solution and the middle portion is in cylindrical shape with lesser diameter than that of the U-shaped portion. The Laboratory Experimental Set-Up for the Sankaranarayanan -Ramasamy (S-R) Crystal Growth Method is simple and user friendly.

4. Salient Features of SR Method

Some of the salient features of SR method are (i) Single crystal with desired orientation is possible at room temperature (ii) With a thin plate as seed, growth of large size crystal is possible. (iii) It is easy to adjust the growth rate as per our need. (iv) The achievement of solute-crystal conversion efficiency is 100%. (v) It is not necessary to prepare all the solution in a time. After mounting the seed crystal with a small amount of solution the rest can be prepared and transferred separately into the growth ampoule. (vi) Simple experimental set-up offers the feeding of the growth solution at a definite interval. (vii) By changing the ampoule shape it is possible to change the shape of the crystal. (viii) The quality of the crystal is always higher than the crystals grown by conventional methods[8].

5. Importance of Unidirectional Crystals

Unidirectional crystals are the need of the hour for the preparation of technological devices. Unidirectional crystals will have highest conversion efficiency of second harmonic generation (SHG). The SHG efficiency is always highest along the phase-match direction for nonlinear optical crystals. The unidirectional crystal growth method is most suitable for grown crystals along phase-match direction.

Table 1: Recent researches on Growth and Characterization of Technologically Significant Bulk Crystals by Unidirectional Sankaranarayanan -Ramasamy Method

Name of the Grown Crystal	XRD Analytical Data	Technological Importance of the unidirectionally grown Crystal
L-Glutaminium p-Toluenesulfonate [9]	Grown crystal is crystallized in monoclinic crystal system with non-centrosymmetric space group $P2_1$. a = 5.4540 Å, b = 8.8013 Å, c = 30.2297 Å, α = γ = 90º, β = 92.835º, volume V = 1449.32 $Å^3$	Optical transmittance of the unidirectional grown crystal is 7 % higher than conventionally grown crystal. Photoluminescence emission wavelength of the grown crystals are observed at 410 nm and it indicates crystals are in violet fluorescence emission. Surface damage resistance from high power laser is increased by 0.3 GW/cm^2. The grown crystals belong to hard material category. TGA and DSC confirmed the grown crystal is thermally stable up to 165 ºC. The SHG efficiency of grown crystal is 0.8 times that of KDP.
Alanine doped KDP single crystals [10]	Potassium Dihydrogen Phosphate (KDP) single crystal belongs to scelenohedral class of tetragonal crystal system point group I42d, space group 122 with lattice constant a = b = 7.448 Å and c = 6.977 Å	The second harmonic frequency generation efficiency of the grown crystal is found to be 1.44 times that of pure KDP. Alanine doped KDP single crystals can be effectively used as a promising NLO material
Xylenol orange dye added KDP crystals [11]	X-ray diffraction patterns of different mole concentration of dye-doped KDP crystals (0.001M and 0.01 M) were compared with pure KDP crystal. The observed prominent peaks of pure and dye-doped KDP crystals are identical but there are variations in peak intensity.	From PXRD pattern, it is observed that Xylenol orange dye-doped KDP crystal has the same structure as that of pure KDP. UV-NIR analysis reveals that the strong absorption is found around 480 nm for the dye-doped KDP crystal. Thermal properties of dye-doped crystals are not altered and remain the same as that of pure KDP crystal. The dye-doped KDP crystal is mechanically stronger than pure KDP. Xylenol orange dye-doped crystal has low dielectric constant and low dielectric loss as it is compared to pure KDP crystal. LDT values of dye-doped KDP crystals are found to be higher than that of pure KDP that is 0.01M Xylenol orange dye-doped crystal has the value 12.40 GW/cm^2 as compared to 8.59 GW/cm^2 for pure KDP crystal.
L-Glutamic acid hydro bromide [12]	The crystal belonged to the orthorhombic crystal system with space group $P2_12_12_1$. a = 5.361Å, b = 11.738 Å, c = 13.372 Å, α = β = γ = 90^0	DTA studies reveal the thermal stability of the grown crystal up to 200°C. UV–VIS–NIR spectrum shows that the SR grown crystals have cut-off wavelength of 230 nm and is more than 70% transparent in the UV region. By studying the etch pits and the growth pattern analysis, growth hillocks were observed and the as-grown LGluHBr crystals follows step-growth pattern.
Pyridinium 2-carboxylate: 4-nitrophenol (P2C4N)[13]	The grown crystal belongs to triclinic crystal system with the space group of P-1.The obtained lattice parameter values are a=6.1682 (±0.0030)Å; b=7.0708 (±0.0060) Å	The cut-off wavelength and refractive index of the grown crystal is 409 nm and 1.34, respectively. Crystal is thermally stable up to 140°C without any weight loss and phase transition. The photoconductivity measurement demonstrates that the grown crystal possesses positive photoconduction property and it is suitable for optoelectronic device

	;c=14.1921 (±0.0088)Å; α=101.4522 (±0.0639)°; β=92.3696 (±0.0451)°; γ=105.0686 (±0.0678)° and V=582.9667 A^3.	applications. The surface laser damage threshold is 2.9 GW/cm^2. The first-order electronic NLO responses have been numerically estimated for the molecule and the study suggested 11 times superior NLO response compared to the organic urea molecule.
T-Stilbene [14]	The material retains its monoclinic crystal system with the space group of P2$_1$/C. The intense diffraction peaks in the pattern articulate the crystalline perfection of the grown material and (011) plane have maximum intensity than other diffracted planes.	The UV-Visible and photoluminescence studies establish the cutoff wavelength at 247 nm with low absorption in the entire visible region and a photoluminescence emission around 385 nm. The crystal sample exhibits characteristic sharp emissions at 385 and 410 nm. The pulse height spectra and scintillation decay time have been studied by using γ –rays excitation from different radioactive sources. Two exponential decay time components of 19 ns (96%) and 337 ns (4%), respectively were observed.
L-Alaninium p-Toluenesulfonate [15]	Grown crystal belongs to the orthorhombic crystal system with non-centrosymmetric space group P2$_1$2$_1$2$_1$. *a* = 5.622 Å, *b* = 8.571 Å, *c* = 26.028 Å, and volume *V* = 1254.27 Å^3	The lower cut-off wavelength of the grown crystals was found to be 285 nm. The transmittance efficiency of the crystal grown by SR method is 20 percentage points higher than that of the crystal grown by conventional method. The LDT value of the SR-grown crystal has increased by 0.57 GW cm^{-2}. The SHG efficiency of the grown crystal is 2.1 times that of KDP.
Ammonium Nickel Sulfate [16]	It is found to be monoclinicin structure with the space group of *P*2$_1$/*c*.*a* = 6.241 Å, *b* = 12.472 Å, *c* = 9.191 Å, and *β* = 106.88°.	The maximum transmission peak (250–270 nm) reached 69%. Thermo-gravimetric analysis revealed the dehydration temperature of about 95 °C for the grown crystal. Thus it is more thermally stable than NSH crystal which has been shown to have dehydration temperature of 73.3 °C.
4-Hydroxy L-Proline[17]	Crystal belongs to orthorhombic crystal system with the non-centrosymmetric space group P2$_1$2$_1$2$_1$. a = 5.01Å, b = 8.28 Å, c = 14.11Å, α = β = γ = 90° and V = 586 Å$^{3.}$	The optical transmittance of the conventional and SR method grown crystals was found to be 60% and 70% respectively. The optical band gap of the crystal was calculated to be 5.23 eV. The thermal diffusivity of the grown crystals were calculated using photoacoustic measurement and the obtained values were found to be 9.435×10^{-5} and 8.990× 10^{-5} m^2/s respectively. The grown crystal is thermally stable up to 265 °C. SHG efficiency is found to be 1.3 times that of KDP material.
Ammonium D,L-Tartrate[18]	The crystal belongs to the orthorhombic system with space group P2$_1$2$_1$2$_1$. *a* = 7.60 Å, *b* = 7.79 Å, *c* = 11.00 Å and volume *V* = 651 Å3	Powder SHG efficiency of was found to be 2.5 times that of standard KDP. The SR method grown crystal has higher LDT value (78 mJ/cm^2) than conventionally (60 mJ/cm^2) grown crystal. The UV–vis NIR transmission has been measured and the observed value of SR method grown AMT crystal is 20% higher than that of conventional slow evaporation method grown crystal.
L-Threonine doped KDP[19]	The crystal system was confirmed to be tetragonal with space group I42d. For pure KDP are *a* = *b* = 7.46860, *c* = 6.96517 Å	The optical transparency of SR-grown KDP crystals was found to be enhanced from 76% to 88%. The band gap for pure and LT-doped SR-grown KDP crystals was found to be 5.68 and 5.53 eV, respectively. The dielectric study also confirms that

	with unit cell volume 386.25783 Å^3 and a = b = 7.44785, c = 7.00015 Å with unit cell volume 388.30166 Å^3 for L-Threonine doped KDP crystal	the grown crystals are free from any major defect and the value of dielectric constant was found to be enhanced due to LT doping. All the studies show that the properties of KDP has been enhanced due to LT doping grown by SR method and can be used as replacement of pure KDP crystal for the fabrication of optoelectronic devices
1,3,5-Triphenylbenzene[20]	The material has been crystallized in orthorhombic crystal system with non-centrosymmetric space group of $Pna2_1$.	The grown crystal has low absorption at the entire visible region and the cutoff wavelength was observed at 247 nm. The material was thermally stable up to 320^{o}C. Grown crystal has SHG efficiency 0.7 times that of KDP
Bisthiourea Zinc Chloride (BTZC)[21]	Orthorhombic system with the space group Pn21a. a=5.706Å, b=12.968Å c=12.762 Å	The transmittance of the SR method grown BTZC crystal is 3% higher than the conventional grown crystal. The micro hardness studies reveal high hardness value for SR method BTZC crystal. Dielectric measurements exhibit very low dielectric constant and loss at higher frequencies, indicating less defects in the samples. The laser damage threshold has been measured and the observed value for SR grown BTZC crystal is higher than that of conventional slow evaporation method grown crystal.
L-Arginine 4-nitrophenolate 4-nitro phenol dehydrate[22]	The crystal belongs to the monoclinic crystallographic system with the space group of $P2_1$. a=7.8Å, b=10.37 Å, c=13.83Å, β=98.21°, V=1117.37Å^3	The transmittance study revealed that the SR method grown LAPP crystal has 10% higher optical transparency than SEST method. The Etch Pit Density is less in SR grown crystal compared to the crystal grown by conventional method. The value of photoconductivity is higher in SR method grown crystal.
Tris (glycine) Calcium (II) dichloride[23]	a=14.736Å, b=19.545Å, c=9.053Å.	The laser damage threshold of conventional method grown crystal is 50 mJ and SR method grown crystal is 70 mJ. The hardness values of conventional and SR crystal are 42 and 53 kg/mm^{2}respectively at 70 g of load. The optical transparency of the conventional and SR method grown crystal was found to be 45 % and 60% respectively.
Triglycine Zinc Chloride[24]	TGZC crystal has an orthorhombic crystal system with space group $Pbn2_1$ and cell parameters: a=11.221Å; b=15.260Å; c= 15.573 Å	Study shows 75% of transmission in the entire visible region, which exhibits good optical quality of the grown crystal. The micro hardness measurement shows that the Vickers micro hardness value is 95 Kg/mm^2. The yield strength is found to be 7.95 MPa. The band gap of the TGZC crystal was found to be ~ 5.2 eV. Dielectric constant is found to be maximum at 1 KHz. The SHG was confirmed by the emission of green light radiation.
L-Lysine added ADP[25]	ADP: a= 7.510 A°, b= 7.510 A°, c= 7.564 A° ADP+L-Lysine: a=7.499A°, b=7.499A°, c=7.548 A°	The crystals exhibited good transparency in the entire UV, visible and near IR region. The crystal doped with L-lysine showed lower transparency compared to the pure one. Vickers micro hardness studies showed that L-Lysine added ADP crystal possessed higher hardness than pure ADP crystal regardless the method by which the crystal was grown.
Lithium Sulphanilate	The Crystal belongs to monoclinic structure with	The transmittance of conventional and unidirectional grown crystals are 62% and 69%, respectively, and

Hydrate[26]	space group P21/c. a = 5.3430 Å, b = 7.9700 Å, c = 19.490Å, $\alpha = \gamma = 90°$, $\beta = 94.099°$ and V = 827.8 $Å^3$	the UV cut-off wavelength is found to be 310 nm. The self-defocusing and the nonlinear refraction as a function of intensity were observed in a Z-scan experiment. The magnitudes for the real and imaginary parts of the third-order nonlinear susceptibility $\chi^{(3)}$ (22.23× 10^{-6} esu) were calculated. The n^2 and β values were found to be 4.512×10^{-9} cm^2/W and 0.177×10^{-4} cm/W, respectively. Z scan reveals that unidirectional grown crystal is suitable for NLO applications.
Ninhydrin[27]	a = 11.2404 Å, b = 6.0604 Å, c = 5.7597 Å, $\beta = 99.1^0$, V = 387.41 $Å^3$	The crystal has good transmittance and there is no absorption in the entire visible region of the spectrum. The calculated band gap is 2.47 eV. Its thermal stability has been determined with respect to different temperatures. SHG efficiency of Ninhydrin is 4.8 times higher than that of standard potassium dihydrogen phosphate.
L-Arginine Phosphate Monohydrate[28]	The grown crystal belongs to the monoclinic system having a non-centrosymmetric structure with space group $P2_1$. a = 10.8576 Å, b = 7.9171 Å, c = 7.3192 Å, $\alpha = \gamma = 90^0$, $\beta = 97.980^0$	The high-resolution diffraction curves reveal the presence of a very low density of edge dislocations along the growth direction. It is observed from the micro-hardness studies that the crystal belongs to the soft material category. The observed stable dielectric constant and low loss values indicate that the grown bulk crystal was free from major defects.
α-$NiSO_4\cdot6H_2O$[29]	a= 6.782 Å b= 6.782 Å c= 18.280 Å The grown α-NSH crystalphase is found to be tetragonal with the space group of $P4_12_12$	The XRD analysis proved good crystallinity of the grown crystal. Optical transmission spectrum of the grown crystal showed high transmission efficiency at UV region and high absorption in the other regions making it suitable for use as UV band-pass filter. Thermal analysis of the grown crystal revealed its dehydration temperature about 73 ◦C.
β-Alanine doped Glycine Phosphite[30]	Crystal exhibits monoclinic crystal system. a = 9.804A°, b=8.498A°, c =7.456A°, $\alpha = \gamma = 90°$ and $\beta = 100.83°$.	The laser damage threshold obtained on the β-alanine doped GPI was 35 mJ/cm^2.From SEM analysis it is observed that the surface of β-alanine-GPI appears with one or two microcrystals and pits. The AFM image of 5 μm size has been recorded.
Bibenzyl & Diphenylacetylene composite t-stilbene[31]	The structural parameters were exemplified by the powder X-ray diffractometer. The crystal samples crystallized in monoclinic crystal system.	The characteristic functional groups were illustrated through micro-Raman spectral measurements. The photo emissive properties of the pure and composite crystals have been analyzed through photoluminescence and X-ray excited studies with the excitation of 275 nm. The scintillation characterizations were evolved by X-ray luminescence and scintillation decay time, pulse shape discrimination studies using 137Cs and 252Cf as a radiation sources. The t-stilbene:Bibenzyl composite crystals has the better FOM value than diphenylacetylene composed t-stilbene single crystal

6.Conclusion:

Sankaranarayanan-Ramasamy (SR) method is a bestunidirectional method to grow huge size bulk crystals. By this growth method, single crystals shall be grown in a precise well-ordered Orientational direction by achieving higher solution to crystallization rate. Most of the studies proved that, the SR Method grown Crystals out performed than the Crystals grown by other Conventional Solution growth methods. Physical, Chemical, Electrical, Optical, Thermal and Mechanical properties of the crystals have been enhanced by growing the crystals by SR growth method. SR grown crystals are of much importance in Technological fields such as Electrical, Optical and Photonics industries. The cut and polished ingots are being used in most of the important industrial devices.

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