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Synthesis and Characterization of CdTr single crystals by gel technique

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ABSTRACT

In the present study, single crystals of cadmium tartrate (CdTr) were grown by a simple gel technique using single diffusion method. The optimum growth conditions were optimized by varying various process parameters such as pH of the gel solution, gel concentration, gel setting time, concentration of the reactance. The test tubes were used as crystallization vessels while silica gel as a growth media. Gel was prepared by mixing the solutions tartaric acid ($C_4H_6O_6$), sodium meta silicate (Na_2SiO_3), cadmium chloride ($CdCl_2$) and transferred in glass tube of diameter 2.5cm and 15cm in length. The mouth of tube was covered by cotton plug and kept it for the setting. After setting the gel, it was left for aging. After two days duration the supernatant tartaric acid ($C_4H_6O_6$) of 1M concentration was poured over the set gel by using pipette then it was kept undisturbed by covering the cotton plug on the mouth of tubes. After 48 hours of pouring the supernatant, the small nucleation growth was observed at below the interface of gel. The round shaped single good quality large yellowish crystals were grown in 45 days. These crystals were characterized by using X-ray diffraction and FTIR analysis.

Keywords: Gel technique, Cadmium Tartrate crystal, XRD, FTIR.

INTRODUCTION

Scientifically and technologically crystal growth and characterization have became an interested research area in the past decades. All basic solid materials are made up of single crystals and they are backbone of the modern technology. The influence of single crystal is noticed in the semiconductors, optics and acoustics, in various medical applications and in jewellary industries [1-4]. Now a day, various crystals are used in electronic industry for controlling the frequency of radio waves, optical property in polarizing microscopes, in microwave communication, in digital telephonic instrumentation, in wireless and optical communication, in electronic and photonic devices [5-9].

Several researchers were found out the different techniques to grow the various types of crystals. The high quality ANI single crystals on SiC substrate were grown by sublimation method [10], lead molybdates crystals and calcium fluoride single crystals were grown by Bridgeman and Czochralski methods [11,12], germanium dioxide (GeO_2), rare earth aluminum borate, $SrMO_{0.93}O_3$, magnesium silicate Peroviskite cadmium sulphide (CdS) crystals, calcium tungustate, transparent piezoelectric crystals of α-$GaPO_4$ were grown by flux method [13-16] whereas piezoelectric quartz crystals, nitrogen doped ZnO crystals and potassium titanil phosphate (KTP) crystals were grown by the hydrothermal method [17,18]. The Cr-doped and CO-doped CdSe, lead zirconate titanate (PZT) crystal were grown by solution growth method [19, 20] while metals, semiconductors ionic crystals and few organic crystals were grown by melt growth method [21].

A systematic study of crystallization in gels begins with Lissegang's famous discovery of periodic crystallization in gels. This method has gained considerable attention because of its simplicity and effectiveness in growing single crystal of certain compound. This method is an alternative technique to solution growth with controlled diffusion. This growth process is free from convection. This is purifying process, free from thermal strain [22, 23]. Crystal habit of various crystals, grown under different conditions and also by different methods were described by H. E. Buckley [24], P. Hartman [25], K. Kern [26], A. A. Chernor [27], W. K. Burton [28] and J. W. Mullin [29]. The various process parameters such as degree of saturation, type of solvent [30], pH of the gel media [31, 32], presence of impurities [33] and the change in growth temperature also presumably affect significantly the morphology of the crystal [34].

Henisch explained the use of gel to grow the crystals [35]. Chemical reaction, chemical reduction, complex dilution and reduction of solubility are the popular methods of crystal growth in gel. Gel media prevents turbulence and helps in formation of crystal by providing a framework of nucleation site. This media is a unique place due to its characteristics of suppression of nucleation centre. The convection is absent in the gel media and it is alternative method for those substances which are insoluble in water [36-38]. The crystals of sulphates [39], crystals of tartrates [40], crystals of molybdates [41], crystals of barium oxalates [42], crystals of iodates [39] were grown by chemical reaction method. Crystals of selenium, lead, copper were grown by chemical reduction method [43] whereas mercury sulphide crystals were grown by complex dilution method [44]. KDP and ADP crystals were grown by reduction of solubility method [45]. Few researchers reported the growth of the single crystals of strontium tartrate tetrahydrates and trihydrates based on their second harmaonic generation characteristics [46-48], single crystals of lithium-doped strontium tartrate on the surface of the gel [49], single crystals of pure and nickel doped strontium tartrate tetrahydrates in silica gel [50], and single crystals of cadmium tartrate in silica hydrogel [51]. In the present study, single crystals of CdTr were grown by a simple gel technique using diffusion method. The optimum growth conditions for crystals were determined. Optimum conditions were established by varying various process parameters such as pH, concentration gel solution, setting time of the gel solution and concentration of the reactance.

MATERIALS AND METHODS

All chemicals used are of AR grade. The gel was prepared by using $C_4H_6O_6$ and Na_2SiO_3 having different pH values varying from 4.0 to 4.5. The chemicals used for growth of single crystal were $C_4H_6O_6$, Na_2SiO_3 and $CdCl_2$. Different molar mass were tried to determine the optimum growth condition. The test tubes were used as crystallization vessels while silica gel as a growth media.

Gel was prepared by mixing the solutions $C_4H_6O_6$, Na_2SiO_3, $CdCl_2$ in double distilled water and transferred it in a glass tube of diameter 2.5cm and 15cm in length. The mouth of a tube is covered by cotton plug and kept it for the setting. After setting the gel, it was left for aging. After two days duration the supernatant $C_4H_6O_6$ of 1M concentration was poured over the set gel by using pipette then it was kept undisturbed by covering the cotton plug on the mouth of tubes.

For infrared analysis of the crystal, samples are prepared in the form of pallet by taking about 100 mg of the sample, mixed with 0.5 gm of analytical grade dry potassium bromide. The mixture is finely powdered and taken in a die. The die is first evacuated to a pressure of 10^{-3} torr, and subjected to extremely high pressure (about 1200 kg cm^{-3}) about five minutes. This process results into the formation of a fine pallet, which removed from the die and used for scanning the spectrum. Experiments were carried out by changing different concentrations of the reactants.

The chemical reaction inside the gel can be expressed as

$$2C_4H_6O_6 + Na_2SiO_3 \rightarrow 2C_4H_6O_6Na\downarrow + SiO + H_2O$$
$$2C_4H_6O_6Na + CdCl_2 \rightarrow C_4H_6O_6Cd\downarrow + 2NaCl$$

RESULTS AND DISCUSSION

The figure 1 shows the single large yellowish grown crystal of CdTr at the interface while figure 2 shows the round shaped single large good quality yellowish crystal of CdTr with its scaling on a graph paper. The size of the grown CdTr crystal was 9mm×9mm×3mm.

Fig. 1 Crystal of CdTr inside the test tube

Table 1 Experimental detail for the growth of CdTr crystal in silica gel

Constant parameters	Variable parameter	Results
$CdCl_2$(1M), $C_4H_6O_6$(1M), Gel age time 48 hours	pH values 4.0 to 4.5	For 4.0 pH, round shaped single large crystal with yellowish colour was obtained. Size of the crystal was 9mm×9mm×3mm.

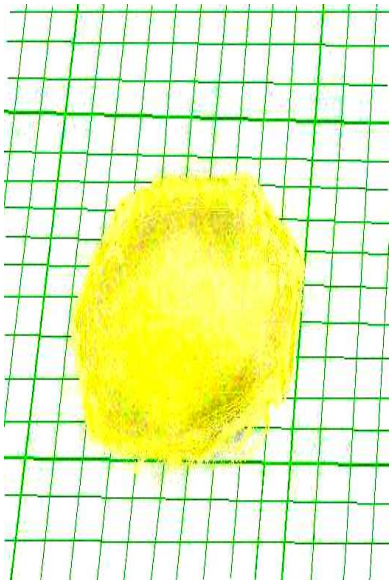

Fig. 2 Single crystal of CdTr

Table 1 shows the experimental detail for the growth of CdTr in silica gel. The various optimum conditions for growing CdTr crystal were established and are given in Table 2. We have changed the process parameters such as pH of the gel and volume of reactants and observe the growth of CdTr. $C_4H_6O_6$ used as upper reactant. Gel age is the time interval between gel and poring of upper reactant.

X- Ray Diffraction Analysis

The crystal structure of the sample compound was studied by powder X-ray diffraction method. The X-ray diffraction was recorded using Miniflex-Rigaku model Japan with CuKα radiation of wavelength λ=1.54056Å. The recorded diffraction pattern of the CdTr crystals is shown in the figure 3.

Determination of Grain size from XRD spectra

From the XRD pattern, it is observed that, each peak has got a finite width. The grain size is determined by measuring the width of the line with highest intensity peak. The grain size can be calculated by using the formula

$$\textbf{Grain size } D = 0.9\,\lambda / \beta\cos\theta$$

Where, β is full width of half maxima in radian and D is grain size of the crystal.

D = 0.9 × 1.54056Å / 0.235× cos (10.8) ˚
= 1.38654/ 0.0040788635
= 344.15 Å
= 34.415 nm

The calculated average grain size is 34.415 nm. The analysis of different diffraction peaks indicates the formation of monoclinic system. The diffraction peaks at 2θ value were measured very carefully and converted into d value using the Bragg's equation putting n=1. By measuring the peak heights above the background in nm and scaling the value up so that the tallest peak has a value of 100.

Table 2 The various optimum conditions of process parameters for growth of CdTr crystals

Sr. No.	Various process parameters	Optimum conditions
01	Density of Na_2SiO_3 solution	1.05 g/cm^3
02	% of Na_2SiO_3 solutions	1.5
03	Concentration of $CdCl_2$	1.0 M
04	Concentration of $C_4H_6O_6$ (supernatant)	1.0 M
05	pH of mixture	4.0
06	Temperature	Room Temp.
07	Gel setting time	72 hours
08	Gel aging time	48 hours
09	Period of crystals growth	7 weeks

The preferential orientation is observed from the XRD data is (-120) indicating maximum growth of the crystal in that direction. Orientations of the crystallites along different h, k and l values are present. The intensity of different peaks could give the relative orientation of a particular h, k and l of plane. In the XRD spectra, the main peaks appeared at various diffraction angles 2θ. The values of 2θ, d values, intensity ratio and their corresponding h, k and l plane were shown in the Table 3. From this table, the observed d values and h, k and l plane were compared with standard data of 2002 JCPDS v. 2.3, 26-0282.

Table 3 Powder diffraction data of CdTr crystal. (λ =1.54056Å)

Observed data values			Standard data values			h k l values
2θ	d value	Intensity	2θ	D value	Intensity	
19.800	4.4801	27	19.364	4.5803	65	0 2 1
21.600	4.1106	100	21.604	4.1102	65	-1 2 0
23.800	3.7354	22	23.771	3.7401	10	-1 0 2
24.200	3.6746	45	24.780	3.5901	14	1 0 1
27.200	3.2757	24	27.080	3.2902	17	-1 2 2
32.200	2.7775	24	32.569	2.7470	13	2 0 0
33.000	2.7120	35	33.152	2.7100	40	1 3 1
34.200	2.6196	51	34.075	2.6290	32	-2 2 2
35.000	2.5615	43	34.742	2.5800	11	-1 0 3
36.600	2.4531	32	36.618	2.4520	20	-2 3 1
37.600	2.3901	29	37.800	2.3781	05	-1 4 2
38.400	2.3421	40	38.575	2.3365	40	-2 0 3
39.400	2.2850	38	39.455	2.2830	40	1 2 2
40.000	2.2521	38	40.320	2.2352	07	2 0 1
41.800	2.1592	51	41.463	2.1760	20	-2 4 1
42.600	2.1204	61	42.256	2.1370	20	0 2 3
43.200	2.0924	27	43.037	2.1000	16	2 2 1
44.200	2.0473	27	44.118	2.0510	14	-2 4 0
46.200	1.9632	79	46.283	1.9600	30	-1 4 3
47.200	1.9240	36	47.123	1.9235	35	-3 2 2
49.400	1.8433	24	49.669	1.8340	17	-2 3 3
50.800	1.7957	77	50.613	1.8020	20	-3 3 1
57.800	1.5938	34	57.557	1.6000	08	3 0 1
58.400	1.5788	36	58.438	1.5781	12	1 4 3
60.800	1.5221	28	60.632	1.5260	02	0 6 3
62.400	1.4869	25	62.585	1.4831	05	1 8 0
63.600	1.4617	43	63.250	1.4690	02	-4 1 1
65.600	1.4219	26	65.597	1.4220	08	3 4 1

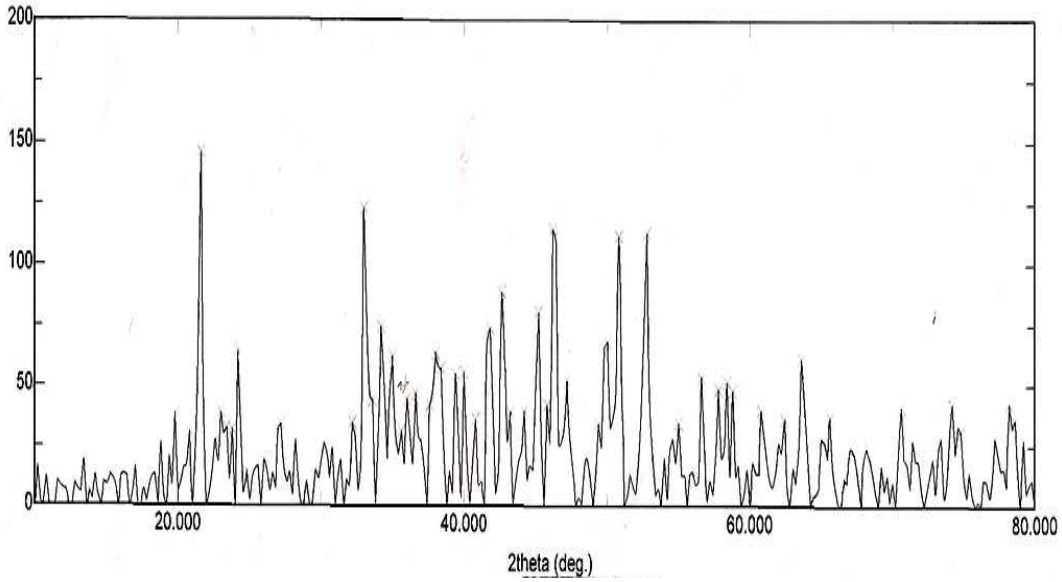

Fig. 3 Powder X-ray diffraction pattern of CdTr Crystal.

FTIR Analysis of CdTr crystal

The FTIR spectrum recorded for CdTr crystal with observed band is shown in the figure 4. The spectrum is scanned in the region 450 to 4000 cm^{-1} using "Perkin Elmer model 783". The –OH stretching frequency of cadmium tartrate appeared at 2924 cm^{-1} confirms the presence of water of crystallization in the crystal [52, 53]. The moderate absorption bands around 3104 to 2916cm^{-1} is probably due to stretching vibration of alkali group. The bands around 1850 to 2800 cm^{-1} may be attributed to hydrogen bonding. The presence of the –C–O– group is indicated by the occurrence of the sharp and an intense band at 1594 cm^{-1} and 1463 cm^{-1} indicates asymmetric –C–H bending. These bands may be assigned respectively to –C–O– asymmetric and symmetric stretching of –C–O–. The bonding mode of water of crystallization overlaps with the new asymmetric –C–O– frequency band. i.e. the region of for the broadness of the absorption around 1593-94 cm^{-1}.

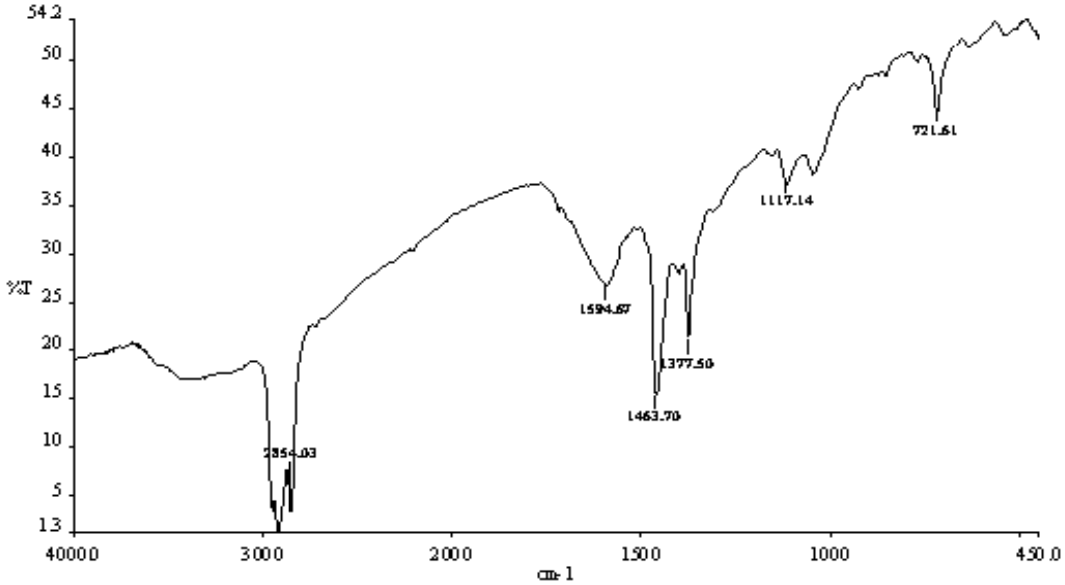

Fig. 4 FTIR Spectrum for CdTr crystal.

The plane bonding of –C–H is assigned by the peak at 1377cm^{-1}. The absorption bands at 1117cm^{-1} are probably due to –O–H bending and –C–OH stretching vibration respectively. It also represents the co-ordinate –C–OH group. The result of FTIR spectra of CdTr crystal with observed band and their assignment are shown in Table 4. It is in good agreement with the structure of the copper tartrate reported by Kirchner [54, 55].

Table 4 The FTIR assignments of CdTr crystal

FTIR peaks cm^{-1}	Intensity	Assignment
2854	Strong, sharp	–OH stretching
3104 – 2916	Weak	C–H stretching of alkali group
1850 – 2800	Weak	Hydrogen bonding
1594	Strong, broad	–C–O– stretching
1463	Strong, sharp	–C–H asymmetric bending
1377	Strong, sharp	–C–H bending
1117	Strong, sharp	–C–OH stretching

CONCLUSION

We have successfully grown CdTr crystal. Single crystals of CdTr were grown by controlled single diffusion of Cd^{2+} using the silica Gel. CdTr is required to maintain the pH of the gel. The process parameters such as temperature affect the growth of CdTr Crystal. Gel growth technique is suitable for growing crystals of CdTr. The round shaped single good quality large yellowish crystals were grown. Size of the grown crystal was 9mm×9mm×3mm. Unit cell parameter values match very well with the reported XRD standard Data values. The FTIR spectrum confirms the formation of CdTr crystal. The CdTr crystal is shining yellowish and quite transparent with good quality.

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