



Studies on growth, optical, electrical and dielectric properties of Strontium and Calcium mixed cadmium oxalate crystals

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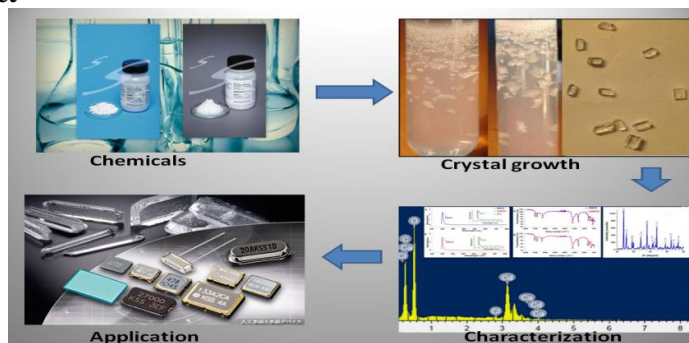
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ABSTRACT

Calcium mixed (CAMCO) and Strontium mixed (SMCO) Cadmium oxalate crystals are grown by gel diffusion method in silica hydro gel by optimizing the growth parameters. Ca^{+2} and Sr^{+2} ions are used to occupy the vacancies of intrinsically available Cd^{+2} ions in the lattice of Cadmium oxalate crystals (ICO). This causes change in morphology of ICO crystals and resulted in the growth of CAMCO and SMCO crystals. Energy dispersive X-ray spectroscopy (EDX) confirmed the presence of Cd^{+2} , Ca^{+2} and Sr^{+2} ions in the lattices of CAMCO and SMCO crystals. Thermo-gravimetric analysis (TGA) of the crystals elucidated the thermal stability up to 950°C and the presence of three water molecules. Fourier transform infrared (FT-IR) spectral studies of the crystals exhibit water of crystallization, carboxyl group and metal-oxygen bonding. Powder X-ray diffraction (P-XRD) studies showed that both CAMCO and SMCO crystals belong to triclinic system. UV-Visible spectroscopic analysis measured the energy gap and insulating behavior of the crystals. Electrical conductivity measurements showed more conductivity in SMCO crystals. CAMCO exhibits higher capacitance than SMCO due to larger dielectric constant. Spectroscopic and structural parameters of both the mixed crystals are compared and analyzed.

Graphical Abstract



High Lights:

Crystals are finest form of solids, used in the study of properties of light, as opto-electronic devices, dielectrics in capacitors and PCBs. Pure, good quality water insoluble crystals were grown using gel

method. We have grown Strontium and Calcium Mixed Cadmium oxalate crystals adopting gel method and their properties were studied.

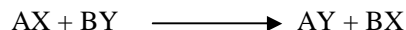
Keywords: Mixed Crystal, Spectrum, Strontium, Calcium, gel, Dielectric

INTRODUCTION

Crystals are more ordered form of solids and appropriate in understanding physical, chemical and optical properties of solids [1]. Mixed crystals are of great importance for their application in the field of semiconductor lasers, diodes and various optoelectronic devices [2]. Certain mixed crystals exhibit exotic properties like piezo-electricity and ferro-eelectricity [3]. Oxalate crystals used as dielectrics in copper clad laminates and in capacitors [4]. In semiconductor industries importance is given to doping and mixing of impurities to intrinsic crystals by various crystallization processes. Nano crystals of Barium doped oxalate crystals show semiconducting properties find their applications in high temperature electronics [4]. Calcium oxalate crystals are used in ceramic glazes. Strontium oxalate crystals are used as auto-extinguishing fibers and in ammunitions [5]. Doping and changing the chemical composition of a crystal can generate new mixed crystals [6]. Here we describe growth and characterization of Ca^{+2} (CAMCO) and Sr^{+2} (SMCO) mixed Cadmium oxalate crystals in single test tube gel diffusion technique. Gel method is most suitable to grow oxalate crystals since they are sparingly soluble in water and decompose before melting. For the grown mixed oxalate crystals thermal, optical and electrical properties were studied.

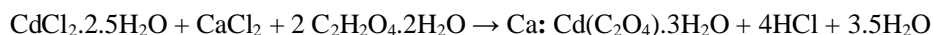
MATERIALS AND METHODS

Mixed crystal can be grown using gel diffusion method, solubility reduction method, the reaction method, chemical reduction method and complex dilution method [2, 3]. Mixed Cadmium oxalate crystals were grown using reaction method, in which single test tube gel diffusion are involved. The general chemical reaction involved in reaction method can be expressed as,



Where A, B are cations and X, Y are the anions [4]. The mixed crystals of Cadmium oxalate are grown by inducing Ca^{2+} and Sr^{2+} ions in the vacancies of intrinsically available Cd^{2+} ions of the parent crystal. Following chemicals are used for growing mixed Cadmium oxalate crystals. Sodium meta silicate (Na_2SiO_3), Oxalic acid ($\text{C}_2\text{H}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$), Strontium chloride ($\text{SrCl}_2 \cdot \text{H}_2\text{O}$), Cadmium chloride ($\text{CdCl}_2 \cdot 2.5\text{H}_2\text{O}$) and Calcium chloride (CaCl_2) of AR grade.

Calcium mixed Cadmium oxalate (CAMCO) and Strontium mixed Cadmium oxalate crystals (SMCO) were grown by single test tube gel diffusion method using silica hydro gel as the growth media at ambient temperature. Na_2SiO_3 (SMS) solution is prepared and diluted to attain specific gravity 1.04. Then the SMS solution is mixed with 0.5 M oxalic acid in a beaker by adding SMS solution drop-wise with constant stirring in the ratio 5:4. The mixed solution are collected in test tubes 9 ml each and allowed to set for 4 days [4- 6]. Then to the test tubes containing gel, the reactant mixtures of 0.5 M CdCl_2 with other reactants 0.5 M CaCl_2 and SrCl_2 in the ratio 5:1 respectively were added. Diffusion of reactants in to the gel resulted the growth of mixed cadmium oxalate crystals [7-9]. The reactions involved in the formation of CAMCO and SMCO crystals are as follows.



Optimum condition for growth of the mixed crystals are recorded in Table 1. Figure 1 illustrates the change in morphology of Cadmium oxalate crystal after mixing with cations Ca^{+2} and Sr^{+2} .

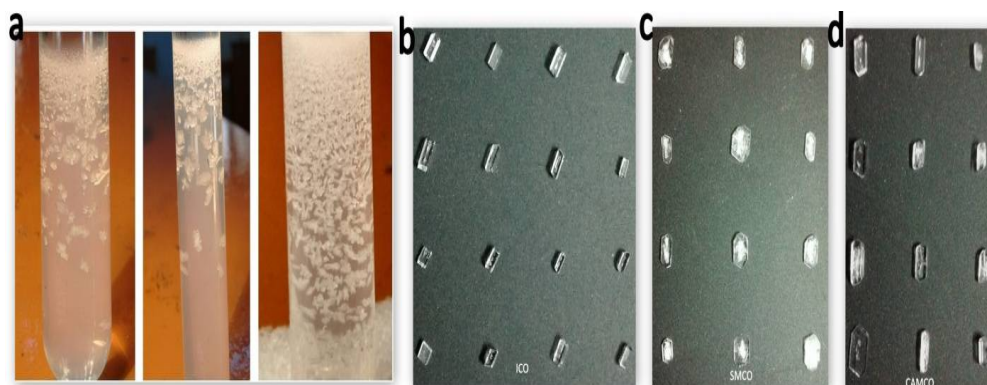


Figure 1. (a) Growth of Cadmium oxalate and mixed oxalate crystals, (b) extracted ICO crystals, (c) extracted SMCO crystals and (d) extracted CAMCO crystals.

Table 1. Optimum growth conditions of SMCO and CAMCO crystals

Parameter	SMCO	CAMCO
Sp. gravity	1.04	1.0425
Gel pH	04.65	05.23
SMS: Oxalic acid	5:4	5:4
Mixture ratio	5.75 : 0.25	5.5 : 0.5
Growth period	15 days	18 days
Chemical formula	Sr : $\text{Cd}(\text{C}_2\text{O}_4) \cdot 3\text{H}_2\text{O}$	Ca : $\text{Cd}(\text{C}_2\text{O}_4) \cdot 3\text{H}_2\text{O}$
Size; $l \times b \times h$ (mm^3)	$5 \times 3 \times 1$	$5.5 \times 3.2 \times 1$
Colour	Colorless	Colorless
Physical appearance	Flat, brittle and transparent	Flat, brittle and transparent

Characterization: Chemical composition and the existence of dopants in the sample crystals were determined using energy dispersive X-ray spectrum (EDX) with the aid of CARL ZIESS FESEM attached with EDS[10, 12]. The decomposition and thermal stability analysis of the crystals were performed using Perkin Elmer Pyris TGA instrument [13- 16]. Temperature was recorded at heating rate of $10^\circ\text{C min}^{-1}$ under N_2 atmosphere. FTIR spectra were recorded using the instrument IR Prestige-21 SHIMADZU for powdered crystalline samples in attenuated total reflectance within wavelength range of 450 to 4000 cm^{-1} [17]. Lattice parameters of mixed oxalate crystals were determined by using powder XRD diffractometer Miniflex 600 Rigaku having X-ray Cu-K alpha of wavelength 1.54 \AA at a scan speed of $1^\circ/\text{minute}$. Band gap energies of CAMCO and SMCO crystals were carried out using UV-Vis spectrophotometer (UV-1800 SCHIMADZU) and the spectrum was recorded in the spectral range of 200 - 1200nm . Further, electrical conductivities of the mixed crystals were measured in the range 0 - 1000 mMho/cm . Dielectric constants of test crystals were measured using Mittal instruments 2151/T-7C calibrated to generate sine wave for frequency 253.88KHz in DSO.

RESULTS AND DISCUSSION

Figure 2 illuminates the electron images of SMCO and CAMCO crystals. EDAX measurements confirmed the existence of impurity cations Ca^{+2} and Sr^{+2} occupying the vacancies of some parental Cd^{+2} ions within the lattices of SMCO and CAMCO crystals (Figure 3). Characteristic peaks observed in the spectra establish the incorporation of elements O, C, Cd and Ca in CAMCO and O, C, Cd and Sr in SMCO crystals respectively. Atomic and weight percentage of elements present in the crystals are listed in Table 2.

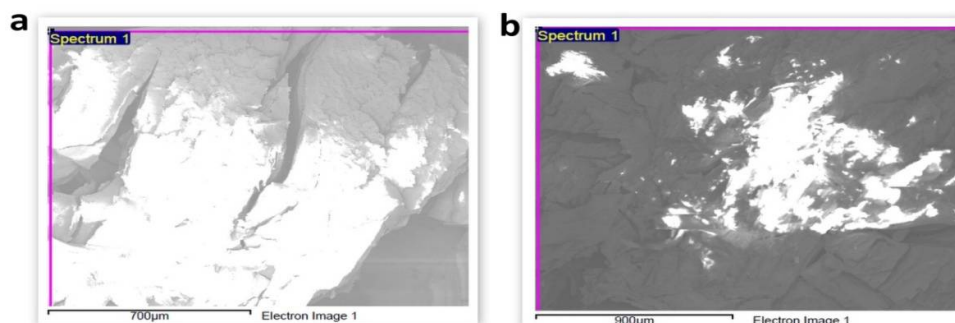


Figure 2. SEM images of (a) SMCO and (b) CAMCO crystals.

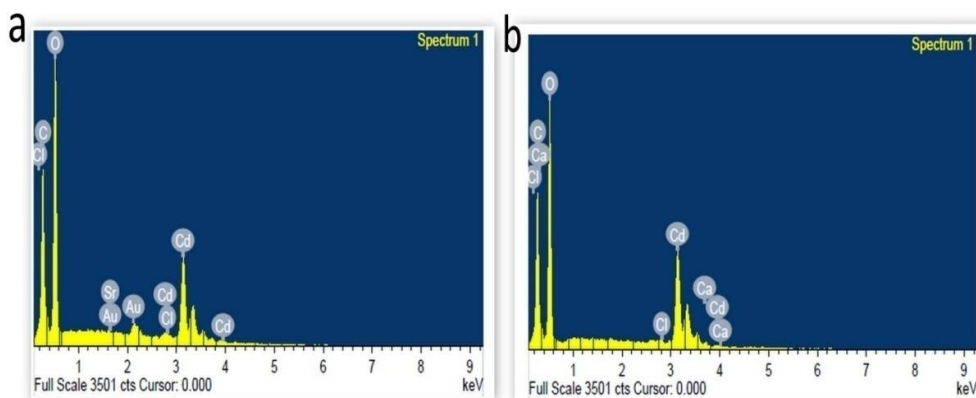


Figure 3. EDAX spectra of (a) SMCO and (b) CAMCO crystals.

The TG plot of SMCO [$\text{Sr}:\text{Cd}(\text{C}_2\text{O}_4)\cdot 3\text{H}_2\text{O}$] revealed that the decomposition occurs in two stages in the temperature range 44–350°C (Figure 4(a)). The first step of thermal decomposition occurs in the temperature range of 44–112°C with measured weight loss of 20.49% (calculated loss: 21.24%) with the loss of three water molecules. The dehydrated SMCO crystal further decomposed at 180–350 °C with a weight loss of 28.077% (calculated loss: 28.318%) losing CO and CO₂ simultaneously. On heating above 350°C the crystal shows stability until 950°C. In the final stage, the material is reduced to its oxide state (>950 °C).

The TG plot of CAMCO [$\text{Ca}:\text{Cd}(\text{C}_2\text{O}_4)\cdot 3\text{H}_2\text{O}$] crystal is shown in Figure 4(b). The decomposition occurs in two stages in the temperature range 51–330°C. The first step of thermal decomposition occurs in the temperature range of 51–127°C with measured weight loss of 20.657% (calculated loss: 21.25%) with the loss of three water molecules. The second stage of decomposition occurs in the temperature range of 227 °C - 330 °C with a weight loss of 27.775% (calculated loss: 28.325%)

loosing CO and CO₂ simultaneously. On heating above 330°C the crystal shows stability until 950°C. TG results of SMCO and CAMCO crystals are listed in Table 3.

The differential scanning calorimetric curves (DSC), (Figure 4(c) and Figure 4(d)) demonstrated the occurrence of small endothermic process around 90–140 °C with a broad endothermic peak at 110.29 °C, which is assigned as the melting point of the crystals.

Table 2. Chemical composition of SMCO and CAMCO crystals.

Crystal	Elements present	Weight%	Atomic%
SMCO	Cd	36.20	06.90
	Sr	00.2	00.05
	O	46.19	6205
	C	17.35	31.00
	Total	100	100
CAMCO	Cd	41.77	08.55
	Ca	00.14	00.09
	O	42.18	60.74
	C	15.91	30.62
	Total	100	100

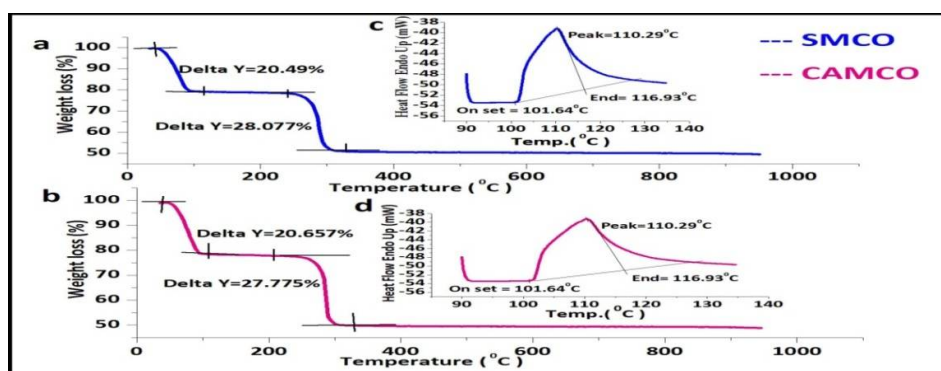


Figure 4. TG plots of (a) SMCO and (b) CAMCO crystals. DSC curves of (c) SMCO and (d) CAMCO crystals.

Table 3. TGA results of CAMCO and SMCO crystals.

Crystal	Calculated weight loss (%)	Observed weight loss (%)	Decomposition temperature (°C)	Molecule decomposed
SMCO	21.24	20.49	44 - 112	3H ₂ O
	28.318	28.077	180 - 350	CO and CO ₂
CAMCO	21.25	20.657	51 - 127	3H ₂ O
	28.325	27.775	227 - 330	CO and CO ₂

FTIR spectra of CAMCO and SMCO crystals are shown in Figure 5 with a wave number range of 500-4500 cm⁻¹. Table 4 lists the FTIR results of CAMCO and SMCO crystals. Both the crystals are associated with similar functional groups. However, there exists a shift in wave numbers of absorption

bands. Fingerprint regions of the spectra confirm the presence of Sr^{+2} and Ca^{+2} ions in the respective crystal lattices.

Both mixed crystals show the presence of an intense broad band (3498 cm^{-1} to 3120 cm^{-1}) are due to the symmetric and asymmetric stretching of O-H group which confirms the water of crystallization [17, 18]. Strong asymmetrical band around 1575 cm^{-1} is attributed due to the C=O stretching in carboxylate ion [19]. The sharp absorption peaks at 1310 cm^{-1} are due to C-C vibrations, C-O stretching [20]. The absorption bands at 780 cm^{-1} – 700 cm^{-1} are due to O-H out of plane bending and metal oxygen (M-O) bonding (Metal = Cd, Sr and Ca). Absorption peaks around 605 cm^{-1} - 560 cm^{-1} correspond to O-M stretching of two crystals respectively.

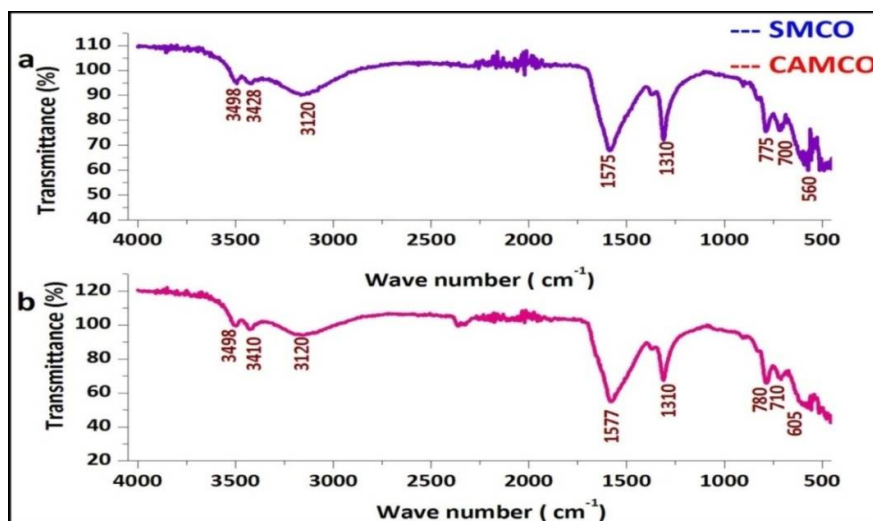


Figure 5. FTIR spectra of (a) SMCO and (b) CAMCO crystals.

Table 4. FTIR results of CAMCO and SMCO crystals

Sl. No.	Band assignments	Wave number cm^{-1}	
		SMCO	CAMCO
01	Symmetric and asymmetric stretching of OH group and water of crystallization	3498	3498
		3428	3410
		3120	3120
02	C=O stretching, O-H bending	1575	1577
03	C-C vibrations, C-O stretching	1310	1310
04	O-H out of plane bending	775 to 700	780 to 710
05	O-M stretching	560	605

Powder XRD patterns of SMCO and CAMCO crystals show well defined peak at specific 2θ values, exhibit high crystalline nature of the grown crystals (Figure 6). Observed XRD pattern of the crystals were indexed using N-TREOR09 program. Obtained d -spacing and the Miller indices are in agreement with the standard values (JCPDS data) [24]. Cell parameters of ICO, SMCO and CAMCO crystals are given in Table 5.

EDX measurements, TG plots, FTIR studies and powder XRD analysis identified the occupation of Ca^{+2} and Sr^{+2} ions in the vacancies of Cd^{+2} ions to form SMCO and CAMCO crystals. The studies also confirm the presence of three water molecules in the lattices of both SMCO and CAMCO and the crystals belong to triclinic system.

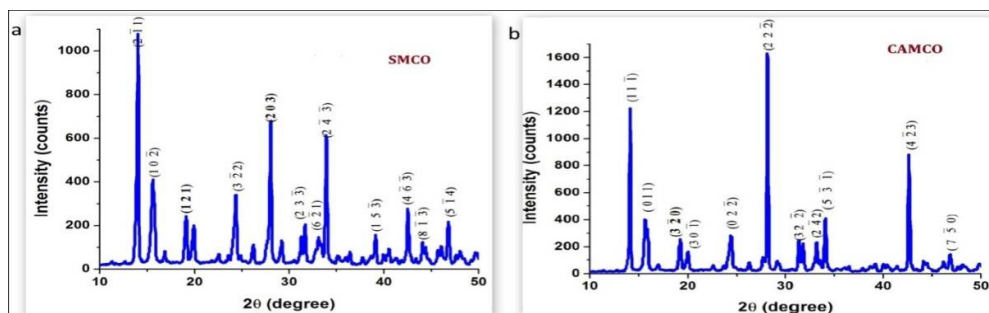


Figure 6. Powder XRD patterns of (a) SMCO and (b) CAMCO crystals.

Table 5. Cell parameters of ICO, SMCO and CAMCO crystals.

Cell parameter	ICO	SMCO	CAMCO
a (Å)	15.4356	18.20618	16.01892
B (Å)	14.0327	15.72032	12.77620
c (Å)	11.5336	11.38376	7.75326
α (°)	103.189	91.747	103.23
β (°)	101.248	102.442	93.126
γ (°)	102.327	115.014	107.885
Space group	P1	P1	P1
Crystal system	Triclinic	Triclinic	Triclinic

Optical properties and energy gap of the mixed oxalate crystals were investigated using UV Visible spectroscopy [21, 22]. Figure 7 shows the variation absorbance of light by the mixed Cadmium oxalate crystals as a function of wavelength. No light photon is absorbed in the visible region, hence crystals become transparent to the visible light. Absorption in the UV region is maximum at 220 nm is 2.435 for SMCO and at 202 nm absorbance is 1.347 for CAMCO crystals respectively. Thus for the SMCO crystal energy gap E_g calculated as

$$E_g \leq \frac{hc}{\lambda_{\min}} \Rightarrow \frac{6.624 \times 10^{-34} \times 3 \times 10^8}{220 \times 10^{-9} \times 1.609 \times 10^{-19}} = 5.61 \text{ eV} \quad (1)$$

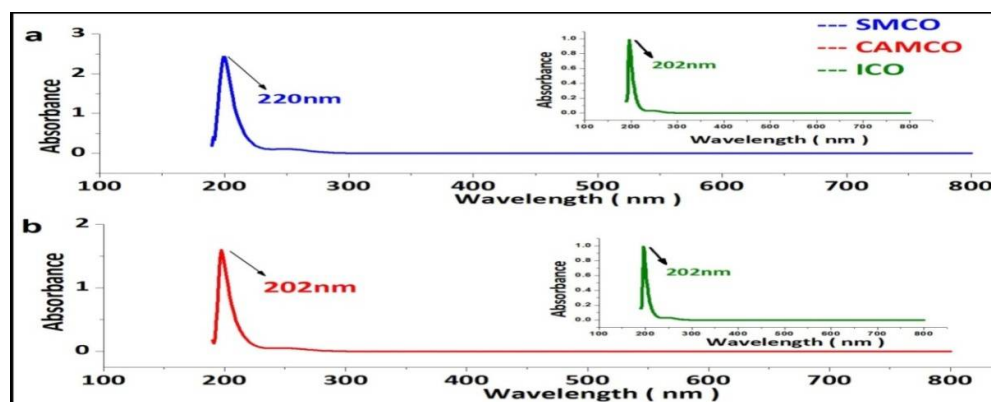


Figure 7. UV- Visible spectra of (a) SMCO and (b) CAMCO crystals.

Further, for ICO and CAMCO crystals absorbance maximum occurs at wavelength of 202 nm give energy gap E_g of 6.11 eV. Due to larger energy gap ICO, SMCO and CAMCO crystals behave as

insulators. The SMCO crystal possesses very small energy gap when compared to CAMCO crystal, which leads to more electrical conductivity in SMCO crystals.

Electrical conductivity of mixed cadmium oxalate crystals were measured by dissolving the crystals in suitable solvents. The solid mixed oxalate crystals are taken in solution form by dissolving 10 mg of test crystal were dissolved in 1.5 N sulphuric acid heated at uniform temperature bath of 80°C for 20 minutes and cooled to lab temperature. The instrument is calibrated to neutralize the conductivity of sulphuric acid [23, 24]. The measured electrical conductivities of ICO, SMCO and CAMCO crystals are shown in Table 6.

Table 6. Electrical conductivity and dielectric measurements of ICO, SMCO and CAMCO crystals.

Crystal	Electrical conductivity mS/cm	Dielectric constant	Refractive index	Capacitance pF
ICO	4.4	34.04	5.83	37.657
SMCO	39.8	14.55	3.81	46.368
CAMCO	35.8	22.42	4.73	51.39

Conductivity of ICO is enhanced remarkably after mixing with Ca⁺² and Sr⁺² ions. SMCO crystal exhibits more conductivity than ICO and CAMCO crystals. This is due to smaller energy gap of SMCO when compared to other two crystals. The above results show that if mixed cadmium oxalate crystals are used as dielectrics in printed circuit boards, electronic and microelectronic devices, then they would provide better electrical and physical properties.

Dielectric constants of mixed crystals were measured for the crystal pellets of thickness approximately 1mm and area attained to the dimension of gold plated dielectric cell. Capacitances (C) of the mixed crystals were measured by comparing the voltage developed across them with the standard capacitor of capacitance 112pF [25, 26]. Capacitance of air C_o is measured for the thickness (d) and area (A) of the pellet using the relation

$$C_o = \frac{\epsilon_o A}{d} \quad (2)$$

Where, ϵ_o is absolute permittivity of free space.

Dielectric constant of the mixed crystal is measured with the relation,

$$\epsilon_r = \frac{C}{C_o} \quad (3)$$

Where, C is the capacitance of the mixed crystal.

UV- Visible spectroscopic analysis of the crystals established the transparency of ICO, SMCO and CAMCO crystals in the visible range. The refractive index (n) of transparent crystals are measured as $n = \sqrt{\epsilon_r}$. Dielectric constants and refractive indices of ICO, SMCO and CAMCO crystals are recorded in Table 6. SMCO crystal establishes lower values of ϵ_r and n when compared to ICO and CAMCO crystals. This is due to its smaller band gap and higher electrical conductivity.

APPLICATION

The studies on properties of SMCO and CAMCO crystals showed high dielectric constants with more band gap energies and insulating behaviour. These results confirm that if mixed cadmium oxalate crystals are used as dielectrics in printed circuit boards, electronic and microelectronic devices, then

they would provide better electrical and physical properties. The mixed crystals are also suitable in crystal oscillators and microwave applications.

CONCLUSIONS

SMCO and CAMCO crystals are grown by mixing cations Sr^{+2} and Ca^{+2} with Cd^{+2} ions. Optimum growth condition of the mixed crystals varies with type of cations used, which results a change in the morphology of ICO crystals. EDX analysis confirmed the existence of impurity cations Sr^{+2} and Ca^{+2} matrixes with Cd^{+2} ions to form SMCO and CAMCO crystals. Both the mixed crystals are thermally stable up to 950°C . FTIR spectra of the mixed crystals confirm the presence of water of crystallization, different functional groups and the metal oxygen bond associated with the crystals. The SMCO and CAMCO crystals belong to the triclinic system. Optical studies of the crystals showed that the mixed crystals are highly transparent in the visible region. The SMCO and CAMCO crystals behave as insulators as their energy gaps are 5.61eV and 6.11eV respectively. Conductivity of the intrinsic Cadmium oxalate crystal is enhanced remarkably by mixing cations Ca^{+2} and Sr^{+2} with the parental Cd^{+2} ions. SMCO and CAMCO crystals associated with lower dielectric constants due to their higher conductivities than ICO crystal.

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