Growth and impedance analysis of pure TGAc and dye doped TGAc crystals-enhanced dielectric permittivity for energy-storage devices

P. R. Deepthi¹ · Anu Sukhdev¹ · P. Mohan Kumar¹ · J. Shanthi² · B. C. Hemaraju³

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Abstract

Herein, we delineate the enhancement of the dielectric properties of an anionic dye doped triglycine acetate crystal for the first time. Single crystals of pure triglycine acetate (TGAc) and reactive orange 16 (RO16) dye-doped (0.01, 0.03 mol%) triglycine acetate were synthesized with an intention to enhance the strengths of pure TGAc crystal using slow evaporation process. The crystalline structure and phase purity of the grown crystals were analyzed using Powder XRD studies. The frequency dependence of real and imaginary part of dielectric constant, loss tangent, real and imaginary part of impedance, electrical modulus and ac electrical conductivity have been investigated. The dielectric constant and dielectric loss for the grown crystals, have been found to decrease with increasing frequency. The decrease in permittivity and dielectric loss with an increase in applied field frequency is as per Maxwell–Wagner theory. The Cole–Cole plot implies that the mechanism of conduction is mainly due to bulk resistance. The enhanced dielectric constant of the doped crystals confirms the appropriateness of the developed crystals for energy storage capacitor applications.

Keywords Dye doped crystal · Dielectric properties · Electrical conductivity

1 Introduction

The potential of electrical energy storage is a deciding consideration for various power systems, immobile power systems, hybrid powered vehicles and pulses [1, 2]. Electric power storage is an important aspect of this. In addition, capacitors are needed to develop and can accumulate and then supply tremendous quantities of energy instantly. For military and commercial uses, this kind of ‘pulse control’ is required. Such applications require higher energy, higher power and more efficiency. Dielectric materials could be used to preserve stored energy within the frame as long as an external electrical field polarizes the electron transfers across constituent molecules or atoms. Resizing electronic devices such as condensers, necessitates novel interfacial materials with an exceptionally high relative permittivity,

mostly gained from perovskites based on ferroelectric and relaxor [3].

The present innovation is showing a fast change and its appearance on materials is unbelievable. The innovative advancement to a bigger degree is subject to the improvement of crystal growth. Hence the growth of single crystals has become foreseeable for any advancement in material science research. The electro-optic constant of a material is directly related to its dielectric constant. Such materials with low or high dielectric constant, may be needed depending on applications such as electro-optical modulator and interlayer dielectric material. The complex impedance spectroscopy is capable of studying the electrical properties of the materials because, under the influence of the frequency range applied, it can shed light on the contributions to the grain, grain boundary

✉ P. R. Deepthi, deeptiprasad82@gmail.com | ¹Material Research Centre, Presidency University, Bengaluru, Karnataka, India. ²Department of Physics, Avinashilingam University for Women, Coimbatore, Tamilnadu, India. ³Department of Physics, Malnad College of Engineering, Hassan, Karnataka, India.

and electrode interface [4]. Amino acids are fascinating resources for opto-electronic application, as they contain carboxyl acid ($-\text{COO}$) proton donor group and amino (NH_2) proton acceptor group in them. The smallest amino acids is glycine, found in proteins and is an essential model for theoretical and experimental studies is of distinct importance. New materials for optoelectronic devices can be formed by adding suitable dopants to alter the physical properties of the crystals [5–9].

In single crystal lattice, the optically active chromophores can be triggered by doping them with a dye molecule. Because of its benefits over typical liquid dye lasers, the use of solid matrices that include laser dyes has proved to be a striking substitute to standard liquid dye solutions [10–12]. Hence, Several researchers have been studying the impact of dye presence on the crystal matrix as it has shown impending applications. Pritula et al. [13] examined the prismatic $\{100\}$ growth areas of KDP by the insertion of xlenol orange dye. Reports are available for the superior physical and optical properties of Crystal Violet (CV)-doped KDP crystals and CV-doped potassium acid phthalate crystals compared to pure KDP [14, 15]. Dye doping, such as methyl orange, rhodamine B, amaranth and xlenol green, has strengthened KDP's single crystal SHG output [13, 16]. Increased optical transmittance, mechanical strength and piezoelectric coefficient resulting from doping with crystal violet dye have been reported in ADP crystal [17]. The improved optical, piezoelectrical, mechanical, and nonlinear optical properties of xlenol orange-doped ADP single crystal have been reported [5]. For sunset yellow doped ADP crystals, enhanced optical, mechanical, and piezoelectric properties have been reported [18]. For RO 16 doped KDP crystals, the enriched optical constants, heat resistance, and nonlinear capability have been reported [10]. So far as the current author's knowledge is concerned, no substantial reports on dye-doped TGAc crystals are available. Reactive Orange 16 (RO 16) is an important class of reactive dyes widely used in the textile industry to color cellulosic fabric as well as wool and polyamide fibers because of their brilliant colour and high water solubility. The structure of RO 16 is given in

Fig. 1. These compounds contain one or more azo groups ($-\text{N}=\text{N}-$), mostly linked to benzene or naphthalene rings. It has two sulfonate groups with negative charges in aqueous solution [10, 19, 20]. Keeping these in back drop, we have explored the influence of an anionic organic dye RO16 in the wide frequency range, the impedance, dielectric constant and mechanism of TGAc's conductivity.

2 Materials and methods

Analar grade glycine and concentrated acetic acid (CH_3COOH) have been used for the growth of crystals. In molar ratio of 3:1, the reagents were dissolved in deionized water to get TGAc crystal [7, 8]. The chemical reaction between glycine and acetic acid is as follows.

The grown crystal was dissolved for a second time in dual distilled water, and then recrystallized by normal vaporization. Within 15 days, distinct and flawless crystals of dimension up to $20 \times 15 \times 11 \text{ mm}^3$ were formed.

The saturated solution for TGAc was first formulated in the case of RO 16 doped TGAc crystals (RO-TGAc). Appropriate amounts of RO 16 were added to this solution as dopant. The seed crystals were collected over a six-day duration and kept in the mother solution for extracting crystals of high quality. Transparent and good quality crystals of RO 16 doped TGAc (RO-TGAc) of dimension $18 \times 14 \times 10 \text{ mm}^3$ and $10 \times 7 \times 8 \text{ mm}^3$ were acquired over a three weeks interval. Figure 2 shows the formed crystals. The shape and size of the grown crystals changed considerably with the inclusion of dye, as shown in Fig. 2. This morphological transition is due to the adsorption of dye on the surface of the crystal. This influences the properties of the solution by changing the interfacial surface tension leading to a change in its anisotropic growth rate.

Fig. 1 The structure of reactive orange 16 dye

Fig. 2 The grown crystals of pure TGAc and ROTGAc **Fig. 3** Powder X-ray diffraction pattern of pure TGAc and RO TGAc single crystals

3 Results and discussion

3.1 X-ray diffraction analysis

X-ray diffractogram (XRD) is the powerful tool for the affirmation of crystallinity and phase purity of the grown crystal. Powder X-ray diffraction evaluation was done by the use of PANALYTICAL X-ray diffractometer with a CuK α radiation ($\lambda = 1.5406 \text{ \AA}$). The XRD pattern of the pure TGAc and ROTGAc crystals is shown in Fig. 3. All the

Table 1 The lattice parameters and cell volume for Pure TGAc and ROTGAc crystals

Lattice parameter	Pure TGAc	0.01 mol% ROTGAc	0.03 mol% ROTGAc
a	5.101	5.110	5.112
b	11.970	11.971	11.973
c	5.461	5.531	5.523
α	90	90	90
β	111.767	111.665	111.687
γ	90	90	90
Volume	309.78	314.44	314.11

crystals possess characteristic diffraction peaks. The distinguished peaks ascertained within the optical anomaly sequence of the synthesized crystal include (1 1 0), (1 0 2), (1 2 1), (2 2 0), (2 2 2), (2 0 2), (2 3 1), (1 4 1), (2 4 1) and (1 0 3) although the strength is seen to differ. Doping with RO 16 dye results in the variation of peak intensity but no shift is observed in the position of XRD peaks. When the dye molecules are incorporated into TGAc's periodic crystal lattice, a strain is induced into the system, resulting in the alteration of the lattice periodicity and decrease in crystal symmetry. As can be seen from the XRD patterns, the diffraction peaks get broadened as the dye concentration is increased, suggesting a systematic decrease in the grain size. Also the incorporation of heavy RO 16 dye molecules induces strain in the TGAc lattice, which results in a slight change in the cell parameter values and decrease in peak intensity, indicating loss of crystallinity due to lattice distortion. Table 1 contains the cell parameters for pure TGAc and ROTGAc crystals. This suggests that the crystals retain single phase structure and exhibit almost the same unit cell parameters. A similar finding was observed in literature [21–23]. Both the pure and doped crystals exhibit monoclinic structure. The minor changes in lattice parameters and the

Fig. 4 W–H graph of pure TGAc crystal

Fig. 6 W–H graph of 0.03 mol% ROTGAc crystal

Fig. 5 W–H graph of 0.01 mol% ROTGAc crystal

appearance of additional peaks reveals the inclusion of reactive orange sixteen in Bravais lattice of TGAc. The findings are in smart accord with the literary works for pure TGAc [7, 8].

The lattice strain induced and the crystallite size of the grown crystals are calculated by applying the Williamson–Hall method. The equation used for the analysis is

$$\beta \cos \theta = \frac{K\lambda}{D} + 4\eta \sin \theta \quad (1)$$

where, β is the FWHM of high intensity diffraction peaks, D is the crystallite size, η is the strain, K is the shape factor equal to 0.94 and λ is the wavelength of the radiation (1.54056 Å for CuK α radiation). The W–H plot for Pure TGAc and ROTGAc crystals are shown in Figs. 4, 5 and 6

Table 2 Crystallite size and strain for Pure TGAc and RO TGAc crystals

Sample name	Crystallite size (nm)	Strain
Pure TGAc	199	0.0043
0.01 mol% ROTGAc	179	0.0065
0.03 mol% ROTGAc	175	0.0076

respectively. The strain and the crystallite size are extracted from the slope and the intercept respectively and is given in Table 2. From Table 2, it is clear that there is an increase in the internal strain of doped crystal relative to that of pure crystal. This could be due to the insertion of RO 16 dye in the matrix lattice. The unit cells of the TGAc molecule need to be extended to accommodate the changed molecule, as a result, local stress being introduced into the area around these cells. The prevalence of the variety of these cells within the crystals contributes to a net internal strain within the crystal. An analogous result was observed in the literature for amino acids doped crystals [24–26]. It is also found that the crystallite size decreases with doping concentration. This is well supported by the dielectric studies of the grown crystals discussed in upcoming section of present communication.

3.2 Dielectric constant

3.2.1 Real part of dielectric constant (ϵ')

The real part ϵ' is the volume of energy retained in the substance from the electrical field and the imaginary part ϵ'' is the quantity of energy dissipation in the device while exposed to a particular electric signal. By using the

subsequent relation the real part of dielectric constant is calculated

$$\epsilon' = \frac{Ct}{A\epsilon_0} \quad (2)$$

where C is the capacitance, t is the thickness of pellet, ϵ_0 is the permittivity of free space and A is the pellet area.

The impact of frequency of electric field on the real part of dielectric constant of pure and doped crystals at room temperature is appeared in Fig. 7. It affirms that the dielectric permittivity diminished with increasing frequency and arrives at a steady value, for all the samples under assessment. The dielectric constant increases with an increase in dopant in the sample. Dielectric constant frequency dispersion depends on polarization phenomena of the type Maxwell–Wagner effect refers to the phenomena of charge separation over a considerable distance in the sample at the interfaces. The inter-well hopping (long range hopping) corresponding to the steady-state transport of charge carriers between the electrodes has no impact on polarization. But due to defects/strain near the grain boundary of all the samples, the probability of inter-well hopping is high and it affects the dielectric relaxation largely at low frequencies [27].

This is natural for a dielectric polar material, and it follows Debye's equation. The dipoles obey the field which is applied at low frequency, while not approaching at high frequency. The materials' relative permittivity is owing to the frequency-dependent involvement of the electronic, ionic, dipolar, and space charges polarizations. Polarization of space charges at low frequencies is more dominant, thus the dielectric constant rises unusually [6]. Frequency

upsurges to a level at which space charge couldn't withstand the applied field. The dielectric constant is also augmented for the crystals doped. RO dye has an ionic feature that helps to create a perfect bonding network in the lattice of crystals. As a result, the crystal's effective polarization is improved, increasing its dielectric constant.

Because of the phenomenon of space charge polarization close to the grain boundaries, which relies upon the purity and perfection of the material, enormous dielectric constant value in the low frequency zone can happen [28, 29]. The effect from space charging to polarization depends on crystal clarity and quality, when impurities and/or flaws identify possible barriers limiting the carriers' transport. The space charge input thus requires a restricted deportation of carriers until they are halted at a potential breaking point, which is typically a grain boundary or phase [30].

3.2.2 Imaginary part of dielectric constant (ϵ'')

Imaginary part of dielectric constant ϵ'' is the measure of dissipation/loss of electrical energy in the sample. It can be estimated using the following relation [31].

$$\epsilon'' = \frac{Gd}{A\omega\epsilon_0} \quad (3)$$

The imaginary part of dielectric constant with alternating field frequency applied is represented in Fig. 8. Dielectric behavior of the samples can be realized based on the idea of the polarization and hopping mechanism. In the pure and doped crystals, the observed dielectric loss is related like this. The hopping of the electron predominates

Fig. 7 Real part of dielectric constant versus log f for pure TGAc and RO TGAc crystals

Fig. 8 Imaginary part of dielectric constant versus log f for pure TGAc and RO TGAc crystals

at lower frequencies, as the frequency enhances the jumping procedure is ended as the hopping may not fulfill the frequency of the field applied; i.e., if τ be the relaxation time of the dipoles, for lower frequencies $f \ll (2\pi\tau)^{-1}$, the dipoles follow the external electric field, therefore a superior value of ϵ'' . At high frequencies $f \gg (2\pi\tau)^{-1}$, the dipoles by no means follow the applied field, and hence, ϵ'' values do not differ significantly. The high dielectric loss value at lower frequencies also suggests the strongly defect free structure and the presence of shallow band gap traps. The shallow traps influence the transport of lattice carriers and excitons. Charge carriers existing in the sample can migrate for certain distance under the influence of an applied electric field. When these carriers are blocked by electrodes, the space charge region is activated, which contributes to a significant increase in ϵ'' in the direction of high frequencies, which results in the reported relaxation of the entire curve.

3.3 Dielectric loss tangent

A material's dielectric loss tangent ($\tan \delta$) denotes quantitative scattering of electrical energy because of different physical procedures, for example, electrical conduction, dielectric relaxation and nonlinear process loss [32]. Figure 9 displays the graph between dielectric loss factors ($\tan \delta$) and frequency. At lower frequencies, the $\tan \delta$ has been identified to be great because of lattice defects which could be due to space charges. At the point when frequency expanded, the estimation of $\tan \delta$ diminished and its worth was seen as extremely low at higher frequencies for both the crystals. In addition, for RO 16 doped TGAc crystal, the dielectric loss was found to be lower than

Fig. 9 Dielectric loss tangent versus $\log f$ for pure TGAc and RO TGAc crystals

that for pure TGAc crystal at advanced frequencies. The perceived low value of dielectric loss at high frequency region for the material endorses that, the quality of crystallization is good and achieved with less defects [32]. A comparative impact was seen in the collected works when KDP is doped with RO 16 dye [10] and TGS is doped with sunset yellow [33].

The reduction in dielectric loss with expanding frequency is accountable for acceptable organic homogeneity of the developed crystals with minor imperfections and this nature is significant for the production of constituents for photonic and electro-optic gadgets. The capacity to help an electrostatic field while dissipating minimal energy in the form of heat is a significant characteristic of NLO material. The lower the dielectric loss, the more productive a dielectric material is [6].

3.4 A.C conductivity

Figure 10 indicates the frequency variability of σ_{ac} . For the grown crystals, the value of σ_{ac} is independent of frequency to low frequency regime whereas, it is found to depend on high frequency regime. Relaxation phenomenon caused by disorder of cations between adjacent locations, mobile charge carriers, and the occurrence of space charge carriers are the factors responsible for the rise in σ_{ac} with the increase in frequency. Whenever a charge carrier hops from the current position to the different site, it will be in a displacement state between two potential well energy minima. The curves are approaching each other to the high frequency that obeys the equation of power law by Jonscher [34].

The power law is given by

Fig. 10 AC conductivity versus $\log f$ for pure TGAc and RO TGAc crystals

Fig. 11 Jonscher plot for pure TGAc crystal

Fig. 13 Jonscher plot for 0.03 mol% ROTGAc crystal

Fig. 12 Jonscher plot for 0.01 mol% ROTGAc crystal

Table 3 Fitting parameters obtained from the Jonscher power law and the value of σ_{dc} for Pure TGAc and RO TGAc crystals

Sample name	σ_{dc} ($\Omega^{-1} \text{ m}^{-1}$)	Transition frequency (Hz)	A ($\text{S m}^{-1} \text{ rad}^{-n}$)	n
Pure TGAc	3.55×10^{-6}	2.34×10^6	0.15×10^{-9}	0.88
0.01 mol% ROTGAc	5.46×10^{-5}	3.09×10^6	4.16×10^{-9}	0.65
0.03 mol% ROTGAc	1.91×10^{-4}	3.38×10^6	7.07×10^{-9}	0.61

suggests that, charge carriers takes a translational motion with a sudden hopping. It can be seen that the value of strength of polarizabilities (A) of pure and doped samples are in accordance with the dielectric results.

$$\sigma_{ac} = Af^n \quad (4)$$

where n and A are constants. The larger value of n ($n > 1$) indicates the contribution of limited bouncing without the species leaving the area and little estimation of ($n < 1$) proposes the translational movement with abrupt jumping. The transition frequency from σ_{dc} to σ_{ac} is alluded as bouncing frequency which indicates the transition from band conduction to polaron hopping conduction [35]. It is also observed that, beyond this hopping frequency, the value of σ_{ac} increases significantly. The Jonscher plots for pure TGAc, 0.01 mol% RO-TGAc and 0.03 mol% RO-TGAc are shown in Figs. 11, 12 and 13 respectively. The calculated values of n and A are given in Table 3. From the Table 3, it is clear that, for all the grown samples $n < 1$. This

3.5 Complex impedance analysis

Using complex modulus spectroscopic analysis, the dielectric information got at room temperature is additionally contemplated. This is a significant apparatus for analyzing the relaxation phenomenon in conducting materials for determining the effects of cathode polarization. Impedance spectroscopy's essential property is its exclusive ability to differentiate the various steps in an ion conducting process including detailed surface and bulk properties information. The real and imaginary parts of the complex impedance is evaluated using the values of capacitance (C_p) and conductance (G), by applying the following standard relations [36, 37]:

$$Z' = \frac{G}{(G^2 + \omega^2 C_p^2)} \quad (5)$$

$$Z'' = \frac{\omega C_p}{(G^2 + \omega^2 C_p^2)} \quad (6)$$

The frequency dependent real (Z') and imaginary (Z'') amount of impedance spectra of all the samples are presented in Figs. 14 and 15 respectively. It is seen that the real impedance part diminishes directly with an expansion in frequency and afterward stays predictable at higher frequencies, proposing an increment in conductivity to the ac. The values of Z' (Fig. 14) appear to converge at greater frequency for all the grown crystals. This is due to the loss of space charges due to the degradation of the barrier characteristics of the sample [38] and might be the cause accountable for increasing the conductivity of the material at upper frequencies. The impedance-versus-frequency pattern gives an indicator of expanding conduction with frequency. The Z'' (Fig. 15) of all the grown samples exhibits peak with distinctive frequency maxima identified as relaxation frequency. The patterns seem to fuse together at higher frequencies. Observed relaxation peaks in the Z'' portion of complex impedance are due to the presence of relaxation of space charge connected to charge carriers causing from oxygen vacancies [39]. In general, wherein the material consists of grain, the polarization of the space charge is found to annihilate in the sample [40]. The Z'' also reduces with increased frequency due to increased losses in the sample's resistive component.

The complex electric modulus is correlated to complex dielectric constant by the following relations

Fig. 14 Variation of impedance Z' (real part) with $\log f$ for pure TGAc and RO TGAc crystals

Fig. 15 Variation of impedance Z'' (imaginary part) with $\log f$ for pure TGAc and RO TGAc crystals

$$M' = \frac{\epsilon'}{(\epsilon')^2 + (\epsilon'')^2} \quad (7)$$

$$M'' = \frac{\epsilon''}{(\epsilon')^2 + (\epsilon'')^2} \quad (8)$$

Here M' is the real part and M'' is the imaginary part of complex electric modulus. The pattern of M' with frequency is appeared in Fig. 16. It is obvious from the figure that the M' esteem increments with an expansion in frequency. This is credited because of the impact of space charge polarization in the materials. The distribution observed here is due to the short range mobility of the charge carriers. Figure 17

Fig. 16 Variation of real part of modulus M' with $\log f$ for pure TGAc and RO TGAc crystals

Fig. 17 Variation of imaginary part of modulus M'' with $\log f$ for pure TGAc and RO TGAc crystals

delineates the variation of M'' with frequency, which shows an expansive peak for each sample demonstrating the presence of relaxation mechanism. The shifting of peaks toward a higher frequency with the addition of dye shows the bouncing of charge transporters at higher frequency [41–43]. This hopping was additionally accountable for high-frequency conduction mechanism. Such charge transporters are limited to their potential wells at inferior frequencies [44]. The relaxation time is calculated from Z'' versus $\log f$ and M'' versus $\log f$ graphs and is specified in Table 4. It is obvious from the table that, in both the cases, the relaxation time are approximately matching.

3.6 Cole–Cole plot

In addition to knowing the conduction type; e.g. either owing to grain effect or grain limit effect, the intricate impedance trace was drawn between actual impedance (Z') and imaginative impedance (Z''). A Cole–Cole trace or Nyquist plot usually consists of twofold back to back

Table 4 Comparison of relaxation time from Z'' versus $\log f$ and M'' versus $\log f$

Sample name	From Z'' versus $\log f$ graph		From M'' versus $\log f$ graph	
	f_r (Hz)	Relaxation time (μ s)	f_r (Hz)	Relaxation time (μ s)
Pure TGAc	371	429.20	323	492.99
0.01 mol% ROTGAc	2089	76.22	2137.9	74.48
0.03 mol% ROTGAc	3000	53.07	3090	51.53

crescents: first part is because of low frequency component of the grain limit and the second, because of high frequency grain or mass properties of the materials. The grown crystals' cole–cole plots are depicted in Fig. 18. It is noted that only one semicircular arc, corresponding to the high grain resistance, is observed for all of the grown samples. A similar behavior is observed in literature for doped ADP crystals [45]. The high value of the resistance to grain is due to the disordered atomic structure near the border of the grain. It is also noted that with an expansion in color density, the radius of the semicircular arc decreases due to the decrease in the resistance resulting from the decrease in grain size by an intensification in color. The increased resistance of grain with dye doping also results in a decrease in relaxation rate. As the dye content increases in the doped TGAc sample, the focuses of the crescent bends to inferior Z' value. It suggests a relaxation effect in the grown crystals that is non Debye-type.

3.7 Complex admittance analysis

In addition to the impedance spectroscopy, the complex admittance spectroscopy is also studied for the developed crystals. The complex admittance spectroscopy is useful for the interlayer formation at electro-electrolyte interface and the charge carrier movement. This spectroscopy consists of capacitance and conductivity as a function of temperature or frequency [46, 47]. The variation of the C and G with the frequency for pure and RO doped TGAc crystals is given in Figs. 19 and 20 respectively. At low frequencies, charges for neutral impurity defects are more readily dispersed, so that defects closest to the positive side of the field are negatively charged while defects closest to the negative side of the field are positively charged. When

Fig. 18 Cole–Cole plot of RO 16 doped TGAc crystals with the inset pure TGAc

Fig. 19 Capacitance versus log f of for pure TGAc and RO TGAc crystals

Fig. 20 Conductance versus log f of for pure TGAc and RO TGAc crystals

the frequency increases, the capacitance decreases to the same level, as the defect charges no longer have time to change in response to the applied voltage.

4 Conclusion

A novel additive, RO16 dye has been utilized in the present investigation, to enhance the dielectric properties of tri-glycine acetate crystals. Single crystals of pure TGAc and RO16-doped TGAc crystals (0.01, 0.03 mol%) were grown using slow evaporation process. The grown crystals exhibits good optical clarity. The nearness of RO16 color in the cross section of TGAc was affirmed by the powder XRD. The powder beam diffraction design illustrates that, the

incorporation of the dopant has no effect on the crystal-line phase. It is found from the dielectric analysis that, dye-added crystal has greater relative permittivity and inferior dielectric loss. Due to the inclusion of RO 16 dye, the grain size decreases, increasing the dielectric constant value. This may be due to a reduction in grain sizes, i.e. an increase in grain surface area, which in turn increases the number of grain boundaries, results in a greater number of dipole formations, as well as an increase in the depletion barrier and, eventually, a high dielectric constant. Variation of dielectric constant and loss ($\tan \delta$) with frequency shows a typical behavior for many ionic solid state systems. Polarizations of the space-charge appear to dominate over ionic or electronic polarizations within the frequency range mentioned. Higher conductivity values at higher frequencies give off an impression that the polarization of the space-charge and hopping processes of the charge carriers. Analysis of impedance spectroscopy affirms the presence of the process of conduction in the grown crystals and is primarily attributable to grain limits.

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Compliance with ethical standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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