Effect of X-ray or γ-ray irradiation on the dielectric properties of calcium tartrate single crystals

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Irradiation of alkali halide crystals with ionizing radiations such as X rays or γ rays is found to affect their physical properties. Studies of such changes in ionic crystals have yielded valuable information regarding the electronic processes taking place in these materials [1–3]. These investigations have also been extended to other solids [4] including ferroelectrics [5].

Work carried out in this laboratory indicates that calcium tartrate is a ferroelectric material with a Curie temperature of 123° C. Irradiation of a ferroelectric material, such as triglycine sulphate, with X rays is found to decrease the value of dielectric constant at low frequencies [6] and also affect the hysteresis loop (it splits into two separate loops) [7]; similar behaviour has been observed in the case of $BaTiO_3$ single crystals [8]. It is felt that an investigation of the dielectric properties, if carried out on calcium tartrate single crystals before and after X-ray or γ-ray irradiation, may give useful information regarding the defects in them. With this end in view, the effect of X-ray or γ-ray irradiation on the dielectric constant and loss of single crystals of calcium tartrate has been studied in the frequency range 10^2–10^7 Hz and in the temperature region 30 to 160° C; the data of such measurements have been reported in this paper. As far as is known to the authors, such data have not been reported till now.

The calcium tartrate single crystals used in the present investigations were grown by the gel technique in our laboratory [9]. The samples had approximate dimensions of 0.8 × 0.5 × 0.1 cm^3.

The samples were exposed to X rays at 30° C for 1 h ($\frac{1}{2}$ h on either side) keeping them at a distance of 2 cm from the target of a Philips X-ray tube run at 35 kV and 10 mA. γ-ray irradiation was carried out using a ^{60}Co source which gives a dose of 78 krad h^{-1}.

The dielectric measurements were taken on a GR 716-type capacitance bridge in the frequency range 10^2 to 10^7 Hz [10]; measurements were taken in the range 10^5 to 10^7 Hz on Marconi-circuit magnification metre type TF 329 G, using a resonance curve principle [4].

Dielectic constant (K) and loss (tan δ) of calcium tartrate samples as a function of frequency at 30° C are shown in Fig. 1. The value of K gradually decreases with frequency and attains a constant value beyond 10^5 Hz; similar behaviour is exhibited by dielectric loss (tan δ); the values of K and tan δ at 30° C and at 10^6 Hz are 7.5 and 0.075 respectively. X-ray or γ-ray irradiation reduces the frequency dependent K value considerably but has less effect on high-frequency dielectric constant. Tan δ values are decreased appreciably on irradiation with X rays or γ rays.

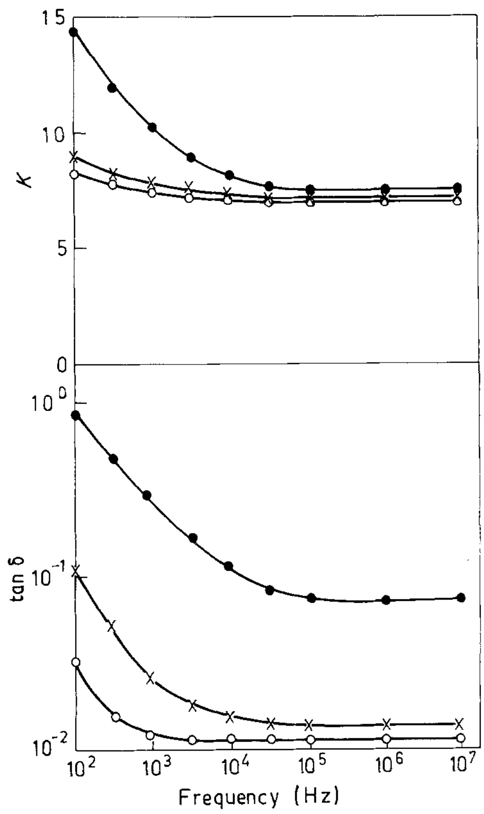

Figure 1 Dielectric constant (K) and loss (tan δ) at 30° C as a function of frequency for calcium tartrate single crystals before (●) and after X-ray (x) or γ-ray (○) irradiation.

The variation of K with temperature at different frequencies for these crystals is presented in Fig. 2. K increases with temperature and decreases with frequency; it goes through a considerable increase at the Curie temperature (123° C). X-ray or γ-ray irradiation reduces the peak value of K significantly at 10^2 Hz, but is found not to have much effect at 10^5 Hz. The peak value of K decreases with the frequency (inset of Fig. 2); the temperature at which K is maximum is independent of frequency. Similar behaviour is exhibited by tan δ (Fig. 3 and inset of Fig. 3). However, it may be noted that γ-ray irradiation reduces the peak value of K and tan δ at 10^2 Hz much more than X-ray irradiation.

A summary of the data of the present work on the calcium tartrate single crystals is given in Table I.

The dielectric constant (K) of the material at low frequencies depends on the electronic, ionic, dipolar orientation and space-charge polarizations. At very low frequencies, all four contributions may be active. In fact, the behaviour of variation of dielectric constant with frequency indicates the relative importance of various types of contributions.

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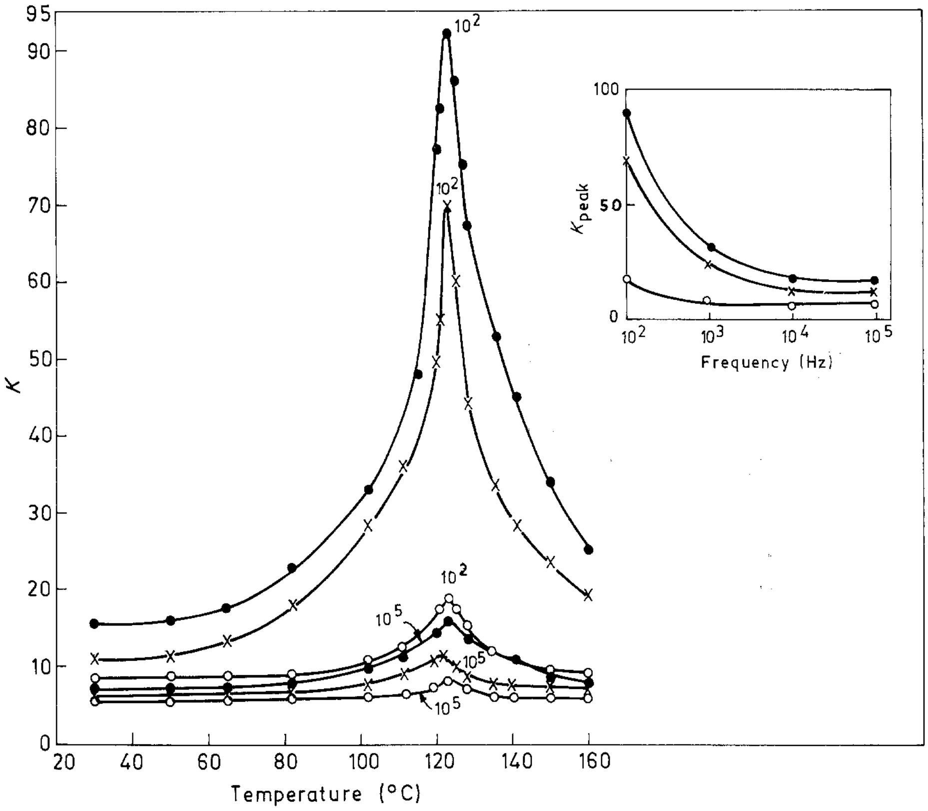

Figure 2 Variation of dielectric constant (K) with temperature at different frequencies for calcium tartrate single crystals before (●) and after X-ray (x) or γ-ray (○) irradiation. Inset: K_{peak} as a function of frequency before and after X-ray or γ-ray irradiation.

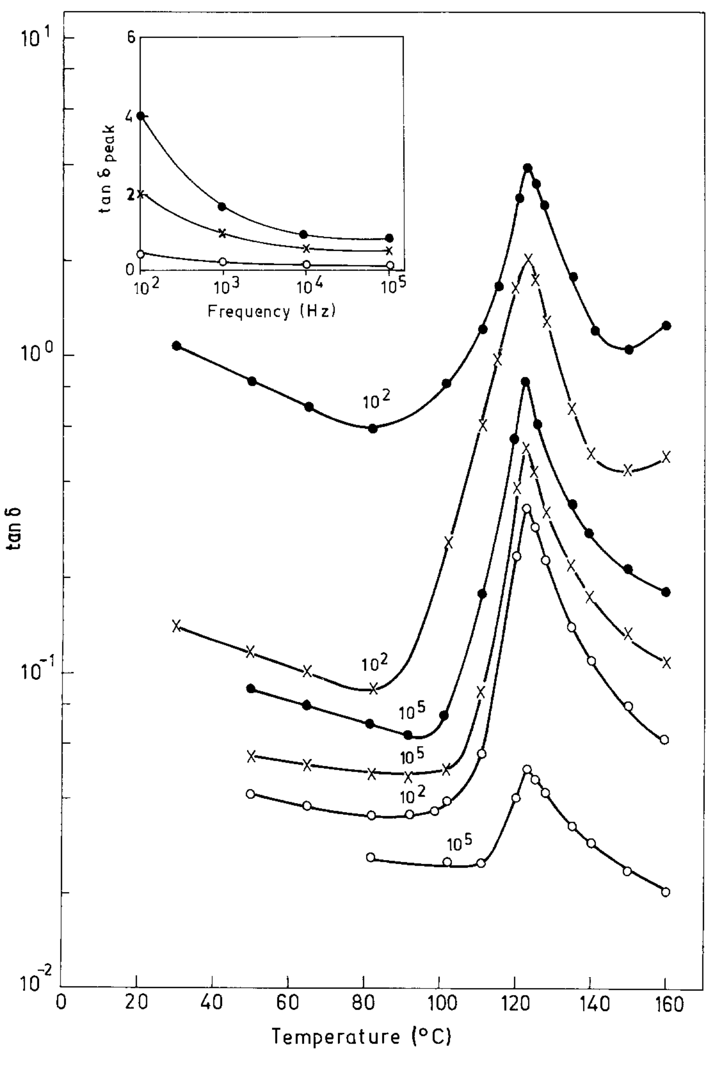

Figure 3 Variation of dielectric loss (tan δ) with temperature at different frequencies for calcium tartrate single crystals before (●) and after X-ray (×) or γ-ray (○) irradiation. Inset: tan δ_{peak} as a function of frequency before and after X-ray or γ-ray irradiation.

TABLE I Summary of data on the dielectric properties of calcium tartrate single crystals

Sample condition	Dielectric constant, K				Dielectric loss			
	at 30° C		at 123° C		at 30° C		at 123° C	
	10^2 Hz	10^5 Hz	10^2 Hz	10^5 Hz	10^2 Hz	10^5 Hz	10^2 Hz	10^5 Hz
As-cleaved	14.3	7.5	92.5	16.0	0.9	0.075	4.0	0.84
X-ray irradiated	9.0	7.3	70.0	11.0	0.11	0.014	2.05	0.52
γ-ray irradiated	8.2	7.0	18.5	8.0	0.032	0.011	0.34	0.05

The contribution of space charge will depend on purity and perfection of the crystals. Its influence is negligible at very low temperatures and is noticeable mainly in the low-frequency region. The dipole-orientational effect in some materials can be seen even up to 10^{10} Hz. The ionic and electronic polarizations always exist below 10^{13} Hz.

Recalling our data, we may ascribe the larger values of dielectric constant and loss exhibited by calcium tartrate at room temperature (30° C) and low frequencies to space-charge polarization due to crystal defects [11]. From the present results it appears that space-charge polarization, which is negligible beyond 10^5 Hz, apparently affects the peak K and tan δ values much more at 10^2 Hz than at 10^5 Hz.

X-ray or γ-ray irradiation of materials produces free electrons in them. The decrease in K and tan δ values of calcium tartrate at low frequencies after irradiation with X rays or γ rays – observed in the present measurements – suggests that these free electrons may be caught up at the charged defects neutralizing some of them, leading to a decrease in space-charge polarization and, hence, the decrease in the dielectric constant and loss. It is also possible that some of the free electrons are tapped at defect regions inside the calcium tartrate crystal producing internal field variations which may clamp the ferroelectric domains thereby decreasing the value of K and tan δ. The reduction in peak K and tan δ values at 10^2Hz in the X-ray or γ-ray irradiated samples may also be due to the same mechanism. Our data indicate that γ-ray irradiation brings out larger decreases in K and tan δ.

In conclusion, it may be mentioned that the present investigations on the dielectric properties of calcium tartrate single crystals at low frequencies are considerably affected by X-ray or γ-ray irradiation.

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