Qatar Univ. Sci. J. (1994), 14 (C) : 100-104

THERMOLUMINESCENCE AND DIELECTRIC PROPERTIES OF AN X-RAY IRRADIATED DOUBLE SULPHATE CRYSTAL

By

H. I. ABD EL-KADER*
Department of Physics, Faculty of Science, University of Qatar, Doha, Qatar
* On leave, Department of Physics, Faculty of Science, Mansoura University, Egypt

الوميض الحرارى وخواص العزل الكهربى لبلورة كبريتات ثنائية مشععة بالأشعة السينية

حامد إبراهيم عبد القادر

فى هذا البحث تم إجراء قياسات الوميض الحرارى والتوصيل الكهربى المتردد σ(ω) لعينات من كبريتات السيزيوم والبوتاسيوم KCs SO4 غير المشععة والمشععة بواسطة الأشعة السينية . وقد استخدم لقياسات الوميض الحرارى جهاز HarShaw model 2000 A+B واستخدم لقياسات المعاوقة جهاز Hioki 3520 . وتوضح النتائج ظهور قمتين منفصلتين فى منحنيات الوميض الحرارى عند جرعات منخفضة من الأشعة السينية ولقد استخدمت اشكال Cole - Cole لتحليل نتائج التوصيل الكهربى المتردد وتم حساب طاقة التنشيط للتوصيل الكهربى المتردد وكذلك لمصايد الوميض الحرارى ومناقشة النتائج. وتوضح نتائج البحث أن بلورات كبريتات السيزيوم والبوتاسيوم يمكن استخدامها فى مجال القياسات الإشعاعية .

Key words: Thermoluminescence, Dielectric properties, Potassium cesium sulphate

ABSTRACT

Thermoluminescence measurements and A. C. conductivity σ(ω) for nonirradiated and x-ray irradiated Potassium Cesium Sulphate, $KCsSO_4$, crystals have been studied. TL reader type Harshaw model 2000 A+B and the impedance meter Hioki 3520 have been used to carry out these measurements. Two separate peaks have been observed TL glow curves at relatively low x-ray doses. The Cole-Cole diagrams have been used to analyse the obtained data. The activation energies for A. C. conductivity and TL trap depth were calculated and the discrepancies were discussed. The obtained results show that $KCsSO_4$ crystals is a good candidate in the field of radiation measurements.

INTRODUCTION

Potassium Cesium Sulphate is one of the double sulphate crystals which is a member of the family of crystals with the general formula $A_1A_2BX_4$ where A_1A_2 are the monovalent ions Li^+, Na^+, K^+, Cs^+, Rb^+, or NH_4^+ and $N_2H_5^+$ groups and BX_4 denotes SO_4^{-2}, SeO_4^{-2} groups. The growing interest in studying the physical properties of double sulphate crystals is due to the fact that they show anomalous behaviour[1-5] at transition temperature (T_c), as well as they undergo an interesting sequence of structural phase transitions[6-9]. The discovery of the ferroelectric, ferroelastic and later on superionic phases divert attention towards studying the physical properties of double sulphate crystals.

These properties are the object of great interest today, due to possibilities of applications in energy sources, IR-detectors, thermal imaging applications[10-11], together with thermoluminescence dosimetry (TLD)[12-13]. Potassium cesium sulphate crystals possess an orthorhombic structure, their lattice parameters obtained from x-ray precession photographs show that the orthorhombic unit cell of $KCsSO_4$ has a_0 = 7.24, b_0 = 12.7, and c_0 = 4.62Å [14]. In this paper, the results of A. C. conductivity and thermoluminescence response of X-ray irradiated KCs101SO4 crystals in the temperature range 300-600K and frequency range 10^2-10^5Hz have been presented and discussed.

EXPERIMENTAL

Crystals of $KCsSO_4$ were prepared by fusion of equimolar amounts of pure K_2SO_4 and Cs_2SO_4. The procedures of the preparation are discussed elsewhere[12) These samples were annealed at 400 °C for one hour, then left to cool gradually, to room temperature, inside the oven.

Irradiation facility

An X-ray machine type TEL-X-OMETER 580 was used for irradiation. The samples to be irradiated were fixed at 7.5 cm from the X-ray tube, the operating voltage of the tube was 20 Kv while the filament current was 8mA. The dose rate at the point of irradiation was 3.83 Rad/min. due to the subjected action of $Cu(K_a)$ radiation. The dose was accumulated by successive exposure ranged from 115 Rad upto 1380 Rad.

Thermoluminescence measurements

Thermoluminescence was measured in temperature range 25-300 °C using a Harshaw 2000 A+B TL-reader. The heating rate was 5 °C/sec. For high precision, measurements were made in duplicates. To avoid oxidation, the system was flushed with pure and dry N_2 gas between and during measurements.

A. C. Conductivity measurements

The material was pressed under pressure of 9.8 x 10^8 Pa to form discs of diameter 12.7 mm and thickness of 1.2 mm. Contacts were made using silver electrodes. The sample was placed in a holder designed to minimize stray capacitance. The dielectric properties were measured using a Hioki 3520 LCR tester. The measurements were performed in the frequency range 0.1-100 KHz. The operating voltage was 0.8 volt. The impedance Z and ϕ can be read directly from the impedance meter, and the values of real and imaginary parts of impedance (Z', Z'') were evaluated. The capacity C and loss factor tan δ were measured and the dielectric constant was calculated.

RESULTS AND DISCUSSION

Potassium cesium sulphate crystals show a reasonable TL response after exposure to X-rays. When the crystal irradiated to a dose of about 115 Rad an increase in the TL peak was observed and at a dose up to 690 Rad the TL peak reaches a maximum, further increase in the dose leads to some decrease in the value of TL-intensity as illustrated in (Fig. 1).

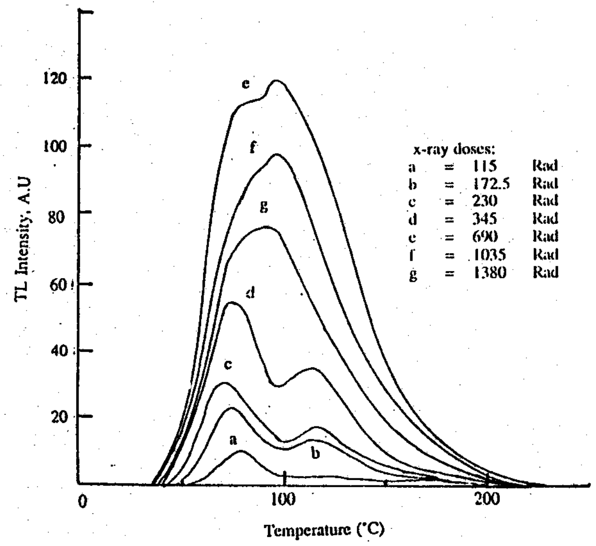

Fig.1. Thermoluminescence glow curves of $KCsSO_4$ at different x-ray doses.

At low doses there are two separate peaks at 75,105°C. The second peak temperature shifts to lower values and the two peaks become singlet[12], this behaviour may be due to defects arising by radiation and located in the layer next to the electrode within half thickness of the specimen[9].

Fig. (2) shows the variation of the second TL peak intensity with irradiation doses,

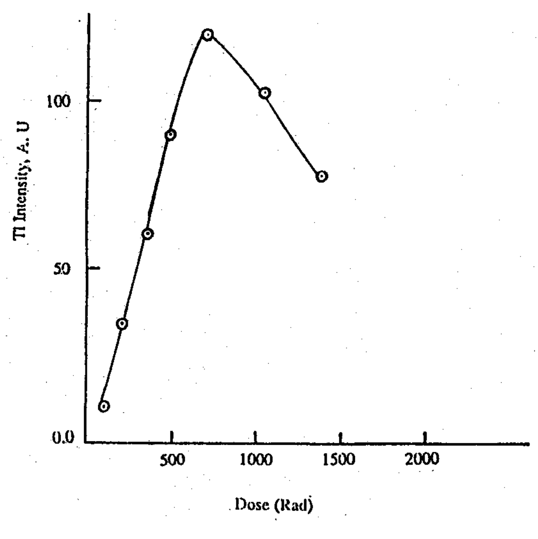

Fig. 2. Variation of TL intensity with x-ray radiation doses.

The figure indicates that the TL peak intensity increases linearly with irradiation doses up to 690 Rad then decreases upon increasing the dose. The decrease of the TL-intensity can be attributed to the radiation damgae of the TL trap in $KCsSO_4$.

The measured impedances of all the samples at various exposure doses of X-ray were analysed using Cole-Cole diagrams[15] for $KCsSO_4$ samples. Fig. (3) shows a typical Cole-Cole diagram for radiated and nonradiated samples.

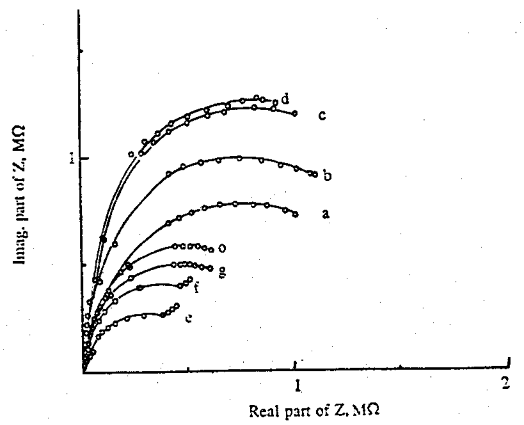

Fig. 3. A complex impedance spectra of $KCsSO_4$ for a range of x-ray doses (Notation as before) and non irradiated sample (o).

The frequency dependence of both the real and imaginary parts (Z', Z'') of the impedance follows semicircles originated from the origin with no overlapping, this means that the surface resistance is Zero and the sample resistance is composed mainly of the bulk resistance which is in parallel with capacitance(C). Extrapolating the high and low frequency limits of the semicircles to intersect with the real axis(Z') give the values of $R_s = 0$ and R_b at different exposure doses. It is also clear from Fig. (3) that the arc diameter increases for the sample irradiated with dose up to 345 Rad followed by a decrease at dose 690 Rad, then upon increasing the dose to 1035, 1380 Rad the arc diameter increases again.

The classical semicircular form can be represented by a simple R-C circuit[16], where

$$C_0 = \lim_{\nu\to\infty}[-2\pi\nu Z'(\nu)]^{-1} \quad (1)$$

is the capacitance of the measured samples whereas the resistance

$$R_0 = \lim_{\nu\to 0} Z'(\nu) \quad (2)$$

is the grain surface resistance . Ionic conductivity of the samples could be estimated using the sample geometry and the DC resistance at different doses. The behaviour of the conductivity with the dose is illustrated in Fig. (4).

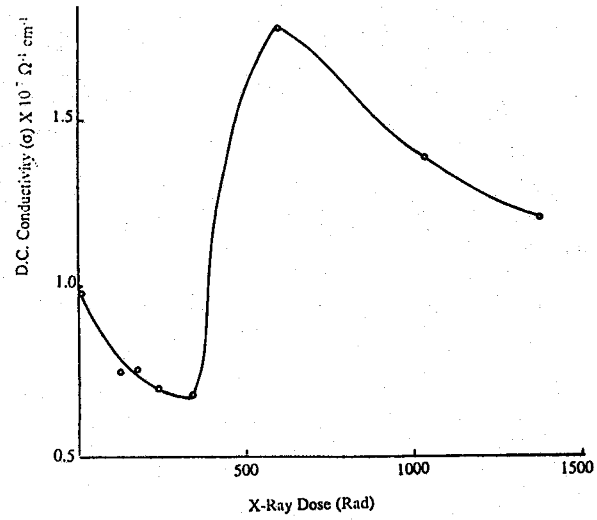

Fig. 4. Conductivity dependence of $KCsSO_4$ with different x-radiation doses.

This figure shows the D. C. conductivity dependence upon X-ray irradiation dose. It is clear from this figure that the value of σ_{DC} changes non-monotonically which, to some extent, characterizes the homogeneity of $KCsSO_4$ crystals in the bulk of these specimens. Fig. (5) shows the relation between log(σ) vs. log(d) which obeys Flower's law[17]

$$\sigma = kd^N$$

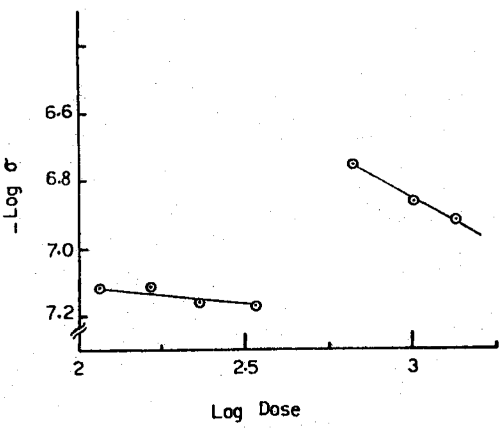

Fig. 5. The relation between log(σ) vs. log(d).

where d is the dose, k and N are constants. It is clear from the figure that there are two different segments. At low doses ($d \leq 345$ Rad) log (σ) decreases linearly with log (d), where σ obeying the empirical formula

$\sigma = 1.35 \times 10^{-7} d^{-0.12}$.

Meanwhile at high doses ($d \geq 690$ Rad) it follows the empirical formula

$\sigma = 1.76 \times 10^{-4} d^{-1.05}$.

Defects are created in $KCsSO_4$ as a result of irradiation. On increasing irradiation dose ($d \leq 345$ Rad) the defects concentration increases and scattering mechanism dominates leading to decrease in conductivity. Further increase of radiation dose (345 >d<690 Rad) electron liberation takes place hence conductivity increases, this is in a good agreement with the obtained TL glow curves which has a maximum TL-response at the same dose (d = 690 Rad), see fig. (2). Upon increasing the irradiation dose, (d >690 Rad), recombination processes takes place hence reducing the conductivity[18]. The variation of A. C. conductivity with frequency shows a good contrast with that of the dielectric constant as shown in (fig. 6 a,b)

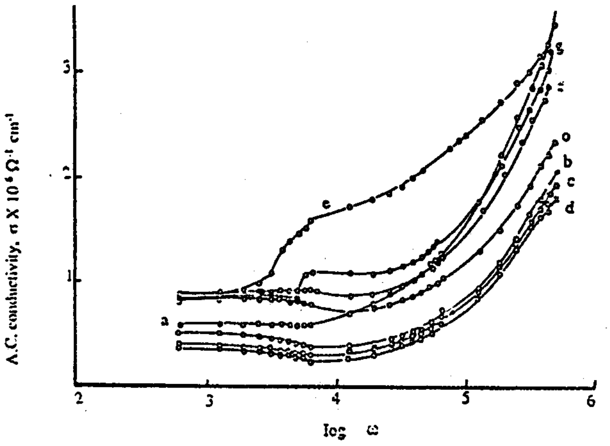

Fig. 6a. The frequency dependence of the conductivity of $KCsSO_4$ with different doses at temperature (T=300K)

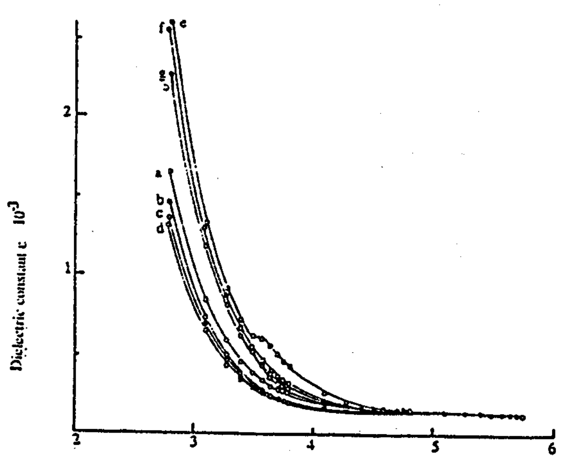

Fig. 6b. Dielectric reponse of $KCsSO_4$ crystals as a function of frequency at different doses. Notation as before.

The conductivity obeyed the empirical law of frequency dependence given by the power low of the form[19]

$$\sigma(\omega) = \sigma_{DC} + \text{Const. } \omega^n \quad (3)$$

where the exponent (n) is in the region 0.4-0.6, this suggests that the possible mechanism of conduction is by hopping.

In the ferroelectric material the theoretical models predict a frequency dependent conductivity either by quantum mechanical tunneling model or hopping over a barrier mechanism[20-21]

The activation energies estimated from AC conductivity measurements for samples irradiated with X-ray doses of 172.5 and 1380 Rad were 3.22 and 2.77 eV, while those estimated from TL measurements were 0.81 and 0.79 eV. The difference in the calculated activation energies using the two techniques can be attributed to self trapping energy at a site[12].

CONCLUSION

From the obtained results we can conclude that the TL-response of $KCsSO_4$ can be used in X-radiation measurements from relatively low doses upto 580 Rad, mean while the dose dependence of dc-conductivity showed nonregular behaviour.

REFERENCES

[1] **Mroz, B., T. Krajewski, T.Breczewski, W. Chomka and D. Semotowicz, 1982.** Ferroelectrics, 42: 459.

[2] **Krajewski, T., T. Breczewski, M. Kassem and B. Mroz, 1984.** Ferroelectrics, 55: 811.

[3] **Kassem, M. E., S. H. Kandil, E. F. El-Wahidy and M. El-Gamal, 1984.** J. Rev. Phys. Appl., 19: 445.

[4] **Kassem, M. E. and S. Hedewy, 1988.** J. Mat. Sci. Lett. 7: 1007.

[5] **Aleksandrov, K. S., I. L. Zherebtsova, I. M. Iskornev, A. I. Kruglik, O. V. Rozanor and I. N. Flerov, 1980.** Sov. Phys. Solid state, 22: 215.

[6] **Mashiyama, H., K. Hasebe, S. Tanisaki, Y. Shiroishi and S. Sawada, 1979.** J. Phys. Soc. Jap., 47: 1198.

[7] **Kihal, B., C. Dugautier, R. Farhi and P. Moch, 1987.** J. Phys. C, 20: 4491.

[8] **Mitsui, T., T. Oka, Y. Shiroishi, M. Takashige, K. Lio and S. Swada, 1975.** J. Phys. Soc. Jap., 39: 845.

[9] **Hiues, M. E. and A. M. Glass, 1977.** Principles and Applications of Ferroelectrics and Related Materials. Claredon Press, Oxford.

[10] **Kassem, M. E., A. M. El-Katib, E. F. El-Wahidy and H. E. Gado, 1987.** J. Mat. Sci. Lett., 6: 507.

[11] **Beerman, H. P., 1977**. Ferroelectrics, 2: 123.

[12] **El-Kolaly, M. A., M. E. Kassem, A. A. Hegazi, L. Z. Ismail** and **L. I. Al-Houty, 1992**. Radiation Effects and Defects in Solids, Vol.124: 437.

[13] **Kassem, M. E., M. A. El-Kolaly** and **Kamal Ismail, 1993**. Material Lett., 16: 102.

[14] **Oye, H., 1964**. Acta. Chem. Scand. 18: 361.

[15] **Jonscher, A. K., 1983**. Dielectric Relaxation in Solids, Chelsea Dielectrics Press Ltd, London.

[16] **Carpentier, J. L., A. Lebrum** and **F. Perdu, 1989**. J. Phys. Chem. Solids, 90: 145.

[17] **Flower, F., 1956**. Proc. Roy. Soc. 23: 6: 646.

[18] **Horowitz, Y. S., 1983**. Thermoluminescence and Thermoluminescent Dosimetry CRC Press Inc Boca Raton, Florida Chap. (2).

[19] **Elliotphil, S. R., 1977**. Mag. 36: 1291.

[20] **Pike, G. E., 1972**. Phys. Rev. B6: 1572.

[21] **Mansingh, A.** and **A. Dhar, 1985**. J. Phys. D. Appl. Phys. 18: 2059.