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THERMOLUMINESCENCE INDUCED BY GAMMA RADIATION IN LiCsSO4 FERROELASTIC CRYSTAL DOPED WITH DIFFERENT HEAVY METALS

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الوهج الوميضي الحراري لبلّورات من كبريتات الليثيوم والسيزيوم الفيرومرنة والمطعمة ببعض العناصر الثقيلة بعد تشعيعها باشعاعات جاما

محمـد عبد الفتـاح القللي

لقد درست خواص الوهج الوميضي لبلورات كبريتات الليثيوم والسيزيوم الفيرومرنة والمطعمة بنسبة ٥ر٪ من العناصر (Os. Tb, Eu and Tl) وكان المدى للحرارة المستخدمة من درجة حرارة الغرفة حتى ٣٠٠° م .

ولقد لوحظ وجود ثلاثة قمم (peaks) – عند درجات حرارة ٧٥ ، ١٢٥ ، ٢٢٥°م – ولقد شملت الدراسة تحليل منحنيات الوهج لوضع كل قمة وارتفاعها عند اختلاف نسب الشوائب وإختلاف الجرعات الاشعاعية لأشعة جاما – ولقد فسرت النتائج على أساس النقاط المعيبة الداخلة في الهيكل التنظيمي للبلورات أو التي قد تحدث بين الجزئيات في الفراغ البلوري – ولقد وجد أن شكل المنحنيات للوهج الحراري يتغير باختلاف نوع العنصر المكون للشوائب المطعم بها تلك البلورات الفيرومرنة – كما نوقشت النتائج بشيء من التفصيل بالإضافة إلى إمكانية استخدام هذه المواد في الأغراض الدوزيمترية وخصوصاً في حالة الحوادث الإشعاعية ، حيث يعتمد ذلك على النهاية الصغرى لمدى حساسية تلك المواد للإشعاعات المؤينة .

Key Words: Thermoluminescence, Gamma radiation, Glow curves, Ferroelastic crystal, Dosimetry.

ABSTRACT

Thermoluminescence (TL) glow curves of lithium cesium sulphate ferroelastic crystals LiCsSO4 doped with 0.5% of Os, Tb, Eu and Tl have been studied in the temperature range from room temperature to 300°C. Three peaks are observed at about 75, 125 and 225°C. The study include also analysis of peak position and peak height of the glow curves with various dopant ions at different gamma radiation doses. The results are interpreted on the basis of point defects introduced interstitially or substitutionally admixtured into the host lattice. In spite of the host material being the same (LiCsSO4), the glow curve structure changes due to variation of dopant materials (Os, Tb, Eu, and Tl). The obtained results are discussed in detail. The applicability of such materials for accident dosimetry depends on the lower limit of detection.

INTRODUCTION

In many cases the properties of imperfect crystals are strongly related to the behaviour of the domain wall in a random medium. This domain wall can co-exist in ferroelastic crystals, because this type of crystals can minimize its free energy by splitting into an optimum number of domains. The energy can be located in the domain wall, and a dense network of domain can provide stress accommodating mechanism(1,2). In a medium with frozen in defects the domain wall deviates from its position and for small concentration of defects the monodomain state is stable against the domain wall formation. The domain wall tends to move if influenced by external or thermal excitations to overcome the barriers. Crystals with general formula MIMIIBX4 in several cases exhibit phase transformation associated with the onset of spontaneous strain in the low and high temperature phase(3-6). This leads in consequence to anomalous changes of their elastic properties (7,8).

The LiCsSO4 crystals undergo a structural phase transition at about 202 K from orthorhombic point group mmm to low temperature monoclinic(7). This is strongly confirmed by calorimetric measurements(8), which demonstrated specific heat anomaly at Tc in the absence of the latent heat and in addition no thermal effect was observed.

Optical, electro- paramagnetic resonance and nuclear magnetic resonance studies of LiCsSO4 crystals have been reported (9-10). The phosphor were made in the period between 1954 and 1960 from pure sample only(12), and are consequently much less sensitive. Systematic studies of various activators and activator combinations led to the development of sensitive phosphors. From these activators Os, Tl, Eu and Tb are generally believed to be those of dosimetric importance. The optimum sensitivity results from addition of 0.5% of the dopant(13-15).

In this paper the thermoluminescence technique was used to find out the effect of gamma radiation induced thermoluminescence in LiCsSO4 doped with 0.5% of Os, Tl, Eu&Tb.

EXPERIMENTAL

The materials used in this work were prepared from high purity Lithium sulphate and Cesium sulphate mixed in equimolar quantities and then heated to 900°C for 4 hours in a platinum crucible. The obtained LiCsSO4 matrix was left to cool to room temperature in the oven. The resulting solid was powdered again and divided into five portions. The first four patches were doped with Tl, Tb, Os and Eu separately. The concentration of each dopant was 0.5% by weight. The fifth part was kept un-doped. Each of the doped samples was mixed thoroughly to form a uniform distribution and then heated at 950°C for 1 hour. After cooling the produced solid samples were ground to 80-120 mesh. The prepared samples were then γ-irradiated with different doses (using Co-60 gamma cell 220 manufactured by Atomic Energy of Canada LTd.). The gamma dose rate was 225 Gy/min.

The thermoluminescence measurements were carried out using a Harshaw 2000 A + B TL reader from room temperature up to 300 °C. The heating rate was 5 °C/sec. To avoid sample oxidation, the system was flushed with pure and dry N2 gas between and during measurements. For precision, each measurement was made twice.

RESULTS AND DISCUSSION

Thermoluminescence of LiCsSO4 has been measured at different gamma radiation exposure doses (0.056, 320 and 675 KGy). The glow curves have been measured in the range from room temperature up to 300 °C. Fig (1) a, b & c represents typical TL glow curves. At low radiation doses ($\approx$ 0.056 KGy) the glow curves show two isolated peaks at 75 and 125°C for non-doped samples. Doped ones show change in glow curve structure in both peak height and peak temperature. This behaviour can be attributed as due to the fact that when the ferroelastic LiCsSO4 crystals are irradiated with gamma radiation the spontaneous strain tensor will change creating an internal stress which tends to diminish the TL responses, the number of populations are decreased by the moderate radiation doses and the TL glow peak becomes triplet with higher TL intensities. By further irradiation ($\approx$ 675 K Gy) the TL trap gains high sensitization and gives high TL response, as is clear from Fig (1-c).

Upon insertion of 0.5% by weight of Tl, the TL response shows two overlaping peaks which increase as the dose increases. In case of Eu, the response shows two faint TL glow peaks. At higher doses this becomes a triplet. In constrast Os

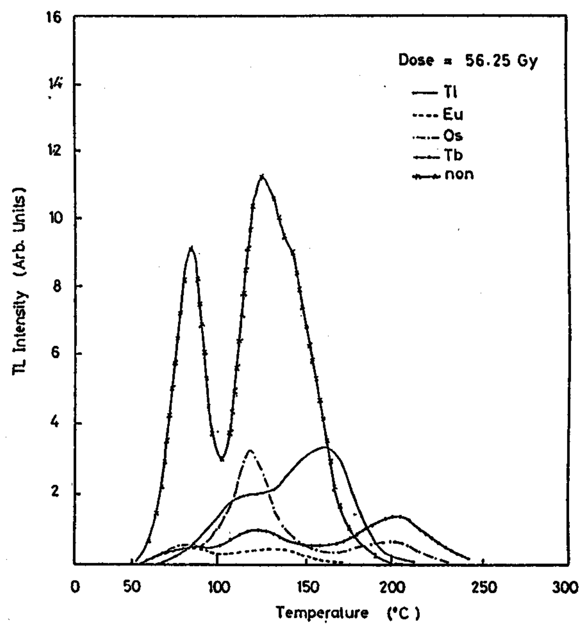
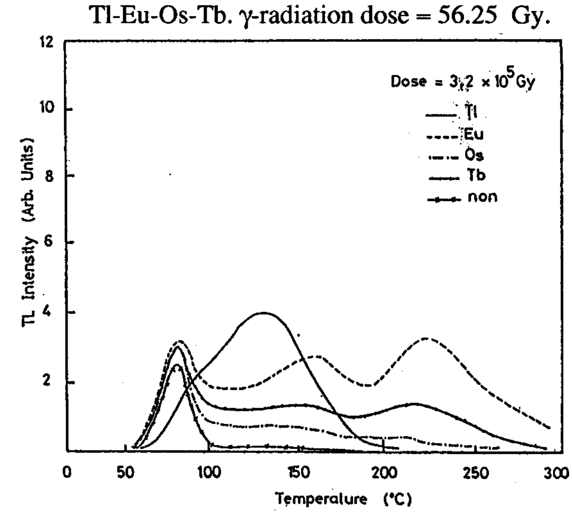


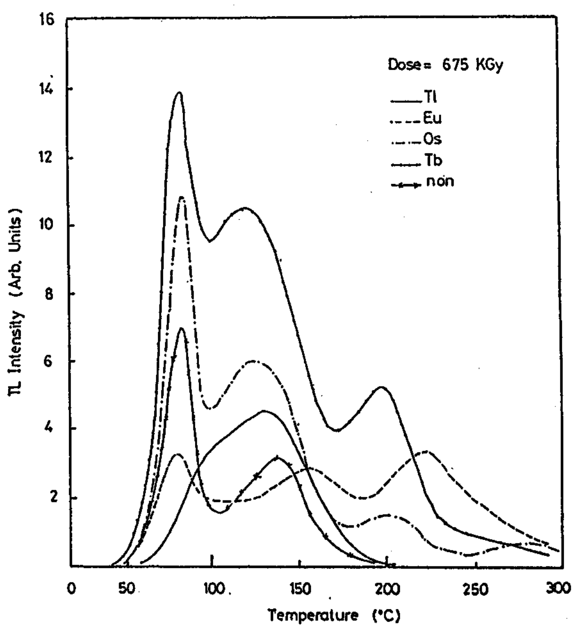
Fig. 1.(a) TL glow curves of γ-irradiated $LiCsSO_4$ crystals both non-doped and doped with 0.5% by weight of Tl-Eu-Os-Tb. γ-radiation dose = 56.25 Gy.



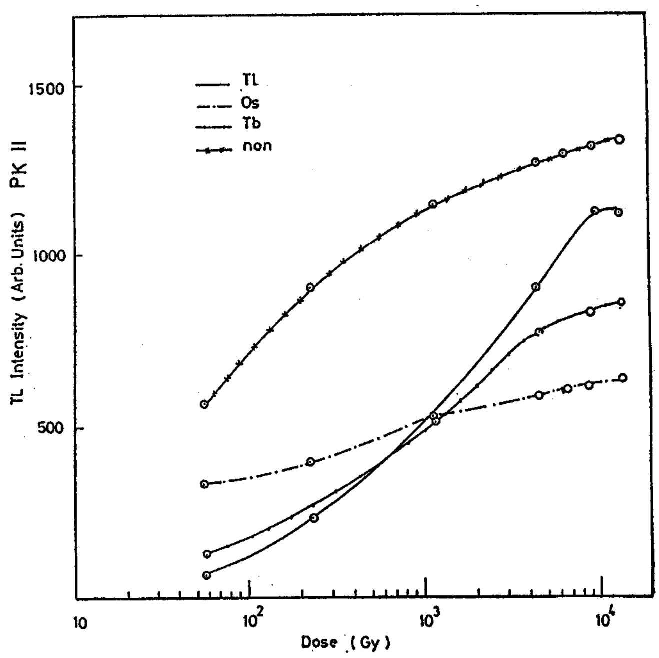
(b) TL glow curves of γ-irradiated $LiCsSO_4$ crystals both non-doped and doped with 0.5% by weight of Tl-Eu-Os-Tb. - γ –radation dose = 320 kGy.

shows a different behaviour in which the TL peak changes slowly with gamma dose till 320 K Gy which is followed by a sharp increase in the TL response at higher doses. With Tb the response shows three faint peaks at low doses. The first peak becomes sharp while the other two change slightly at medium radiation level. The three peaks become sharp and isolated with higher radiation doses.

Fig (2-a) shows the TL intensities of the first peak (PKI) at 75°C versus radiation doses. It is clear from this figure that the TL intensities of LiCsSO4 samples doped with Tb, Os and Eu increase as the radiation doses are increased. In case of non-doped samples this increase goes faster than for doped samples. There are no glow peaks due to Tl in this range of dose. This behaviour can be explained by the fact that the replacement of Cs by Tl breaks the symmetry of the lattice and couples linearly to the order parameter (spontaneous strain tensor). On the other hand Os, Tb and Eu can replace Li or Cs and the influence of defect on TL glow curve depends strongly on the relaxation of the defected cells which tend to order themselves in a way to favour orientation of the order parameter.



(c) TL glow curves of γ-irradiated $LiCsSO_4$ crystals both non-doped and doped with 0.5% by weight of Tl - Eu - Os - Tb. γ-radiation dose = 675 K Gy.



(b) TL response of $LiCsSO_4$ samples both non-doped and doped with 0.5% of Tl, Os, Tb versus g-radiation doses for the second peak (PK II).

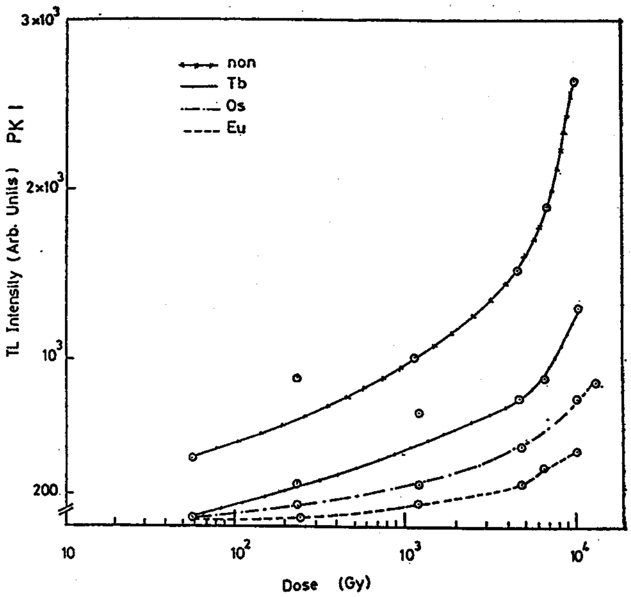


Fig. 2 (a) TL response of $LiCsSO_4$ samples both non-doped and doped with 0.5% of Tb - Os - Eu versus g-radiation dose for the first peak (PK I).

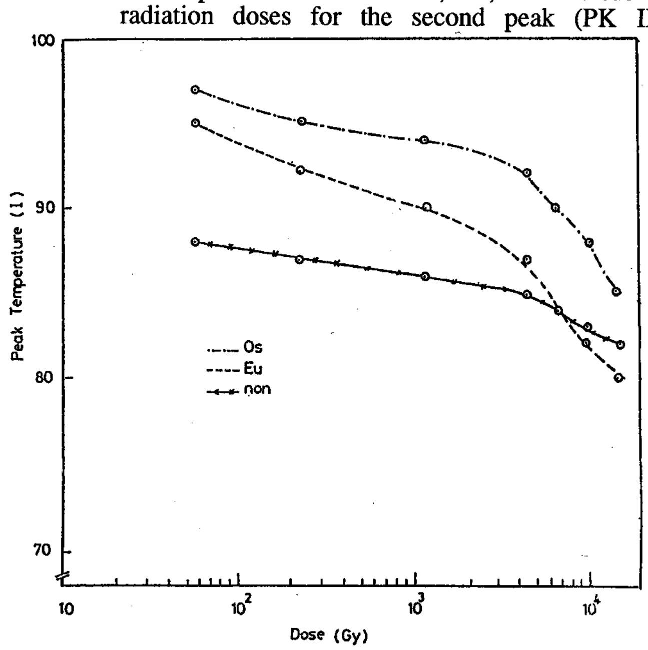


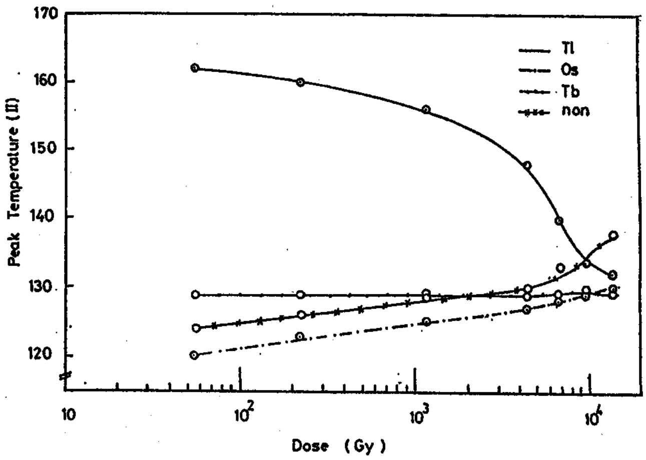
Fig.3. (a) Variation of the TL peak temperature (peak I) of $LiCsSO_4$ samples doped with Os and Eu versus radiation gamma doses.

Fig (2-b) shows also the TL peak intensities (PK II) at (125°C) as a function of radiation doses. It is clear from this figure that non-doped samples as well as Tl, Tb and Os doped samples show an increase with dose. In case of Tl doped samples the increase in TL glow peaks goes faster approximately in the same way as in the non-doped samples. This can be also attributed to the fact that Tl in the LiCsSO4 sample replaces Cs in the lattice. This leads to two phase systems and so, it behaves differently. However, in the other cases Tb, Os and Eu are sited in the interstitial position and these behave similarly.

Fig (3-a) shows variation of peak temperature (The first peak I at 75°C) with radiation dose. It is clear from this figure that peak temperature decreases as radiation dose increases for non-doped samples as well as for Os and Eu doped LiCsSO4 samples. On the other hand, Fig (3-b) shows the variation of peak temperature (Peak II at 150°C) with radiation dose for pure (non-doped) as well as Tl, Os and Tb doped samples. The peak temperature increases slightly with radiation dose. On the contrary, in case of Tl doped samples, the peak temperature decreases with gamma radiation dose. This decrease in the TL intensity may be attributed to the scattering mechanism of the defect with spontaneous strain. Upon increasing radiation doses, the spontaneous strain diminishes, and therefore this defect is dominated.

CONCLUSIONS

From the obtained results one can conclude that heavy metal impurities (Tl, Tb, Os and Eu) play an important role in the thermoluminescence of $LiCsSO_4$ ferroelastic crystals.



(b) Variation of the TL peak temperature (peak II) of $LiCsSO_4$ samples doped with Tl, Os, Tb versus g-radiation doses.

Also, adding 0.5% of Tl by weight to the host lattice gives high TL sensitivity as indicated by the second peak around 150°C up to 1 K Gy gamma radiation dose. Therefore, using these materials in the field of radiation dosimetry is useful in this dose range.

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