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Thermoluminescence Property of γ-Ray Irradiated Cerium Doped Potassium Iodide Single Crystals

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Abstract— This work describes the thermoluminescence (TL) response of cerium doped Potassium iodide crystals irradiated with gamma rays. These crystals gave two glow peaks at 393K and 445K which indicated the nature of traps and shoulder around 370K.All the glow peaks except the peak at 445K attributed to Z-centers, since the intensity of such peaks decreases on F-bleaching. The low temperature peak at 393K has been attributed to Z_1 centers formed on irradiation. The intensity of light emitted dependent on radiation exposure. After one hour of gamma ray irradiation crystals emit visible light around wavelength 430nm when exposed to thermal treatment.

Keywords— KI,Thermoluminescence,TSE,Ce^{3+}

1. INTRODUCTION

Thermoluminescence in alkali halides has now been studied for more than fifty years. It has been fully established that during irradiation of alkali halides defects are trapped at respective trapping centers [1-4]. During annealing of crystals, defects are thermally released from traps and recombine with their complementary defects resulting in the emission of light. This is called TL. The alkali halides seemed to be quite suitable materials to study thermoluminescent processes, because in these either relatively pure (or) doped materials well defined electron and hole trapped centers are induced by irradiation[5,6].It is well known that thermoluminescence property are modified in alkali halide crystal when doped with trivalent impurities. While most of the impurities undergo valence change on irradiation, normally do not undergo a valence change but are known only to perturb the F-Centre configuration, giving rise to a new series of electron excess centers called Z-centers. Recently Z centers in alkali halide crystals are known to offer several attractive features for use as media in holographic information storage[7] and mode locking of high power lasers [8]. Glow curve analysis makes it possible to learn more about the traps, their parameters and nature. We expect that rare earth activated KI crystals can meet the requirements of many applications. So, this paper reports thermoluminescence glow curve of cerium doped KI single crystals grown from melt in Bridgeman furnace and the results of TL emission spectra to identify the nature of emission involved in a glow peak.

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2. EXPERIMENTAL DETAILS

Single crystals of pure KI (99.99% purity) and cerium doped KI (99.99% purity) were grown using the Bridgman Stockbarger technique. Cerium was added in the form of cerium fluoride (Aldrich 99.99% purity). The crystals were grown with three different concentrations of the impurity 1%, 3% and 5% by weight. Samples of size approximately 5x5x1 mm^3 were used for TL studies. The results due to the three concentrations were similar and hence only the results pertaining to a Cerium concentration of 3% by weight are presented and discussed. TL glow were recorded using a PC based TL analyzer (Hitachi make) at a heating rate of 120°C/min the samples were irradiated with an γ- ray source operating at 25kV and 7 mA. TL emission was recorded using Perkin Elmer LS 55 with the excitation slit being closed. Before every experiment, the crystals were annealed at 400°K for half an hour, and then they were quenched to room temperature to ensure homogeneous distribution of impurity and to remove any storage effect.

3. THERMOLUMINESCENCE GLOW CURVE

In order to obtain a better understanding of the TL process, the spectral distribution of TL glow studied for both pure and doped crystals. The TL glow curve of undoped and doped samples after 1 hr of γ-ray irradiation are shown in fig (1) & fig (2) respectively. The TL glow peak of undoped sample centered at 400K and 500K (fig.1). In the case of KI:Ce^{3+} crystals, two glow peaks at 393K and 445K (fig.2) have been observed for 1 hour γ-irradiation. The addition of active ions modifies the obtained TL glow curve for undoped samples The experimental result can be fitted in good approximation by a set of Gaussian curves indicating the presence of two different types of traps characteristic of crystalline structure. The fitting process of experimental data for

KI:Ce^{3+} crystal indicate that the glow curves are composed by same overlapped TL glow peaks presented in undoped samples(fig.2).No new traps related with cerium ions were observed. But in that case, the position, full width at half maximum and then the activation energy were slightly different. The TL glow curve of cerium doped KI is well fitted by set of Gaussian curves(fig. 2).

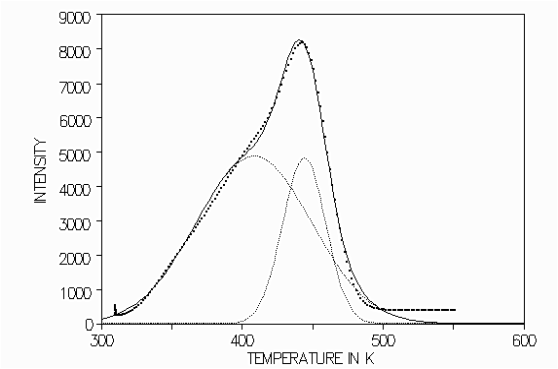

Fig.1 TL glow curve for pure KI γ -ray irradiation1 hr deconvoluted

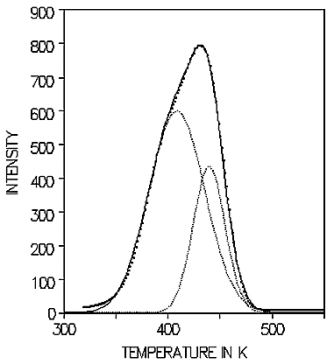

Fig. 2: TL glow curve of KI: Ce^{3+} γ-irradiated for 1 hour

The glow peak at 393K except the peak at 445K have been attributed to F-centers, since the intensity of 445K glow peak decreases on F-bleaching. The behavior of shift of thermo luminescence (TL) glow peak on F-bleaching, it could be concluded that the 393K glow peak attributed to thermal annealing of Z_1 centers formed on irradiation. The thermal stability of Z_1 centers also has been confirmed through optical absorption studies, by observing the removal of the F-band on heating the crystals upto 393K.On F-bleaching, the intensity of F-band is found to diminish accompanying a small increase on the longer wavelength side as shown in fig 2.As discussed in the previous case of KI:Ce^{3+} crystals, correlating the optical absorption and TL results the 393K low temperature glow peak can be safely attributed to Z_1 centers formed on irradiation itself whose concentration is increased on F-bleaching. The shoulder noticed around 370K in KI:Ce^{3+} crystals (which is affected by F-bleaching) may be due to some background impurity probably due to cerium capturing an electron on irradiation and releasing it during thermal excitation as in the case of RbCl:Ca^{2+} studied by Sastry and Sapru [9].The suppression of the 418 and 404K glow peaks(attributed to F-centers) in the initial stage of irradiation confirms this result. The absence of the 370K shoulder and 440K glow peak on higher dose of irradiation may be due to the dominance of F-centers which over shadow the peaks due to impurities. The nature of the centers responsible for the observed glow peaks in the pure and doped crystals could be studied [10] by observing bleaching kinetics of these centers. In pure crystals, bleaching with F-light results in decreases in the intensities of all the glow peaks (not shown in fig) one such typical glow curve of KI:Ce^{3+} crystal γ-irradiated for 1hr and F-bleached for 2-min shown in fig 3.

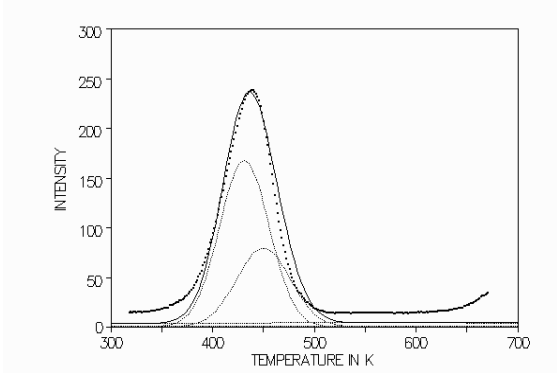

Fig. 3: TL glow curve KI: Ce^{3+} crystals F light bleached 1 min subsequent to γ -ray irradiation for 1 hr

4. THERMOSTIMULATED EMISSION (TSE)

When the sample is heated, the trapped electrons gain activation energy and may return to conduction band. The final step is electron-hole recombination at recombination centers producing light. To study the nature of the recombination centre responsible for TL glow, the emission spectra of 445K glow peak have been recorded. The 430nm emission band observed in KI:Ce^{3+} crystals may be attributed to the recombination of electrons released from different traps with hole centers. The TL emission spectrum of the γ-irradiated KI:Ce^{3+} crystals at 445K exhibit a peak at 430nm and a prominent shoulder at 450nm as shown in fig 4.The prominent shoulder at 450nm can be attributed to the host emission. The emission at 430nm in TSE and indicate that the cerium ions take part in the recombination process. The excitation of the trivalent cerium may be done either optically or by ionizing radiations. Trivalent cerium may be excited to 5d state by ionizing radiation either directly by intra ionic processes within cerium or indirectly. In this case, the excited trivalent cerium emits a 5d-4f photon. The possibility is that the thermally released electrons (from perturbed F-centers) recombine at Ce^{4+} created during the irradiation process. The recombination process leaves the Ce^{3+} (formed after recombination of an e^- with Ce^{4+}) in an

excited state. This Ce^{3+} probably gives the emission observed at 430nm when it returns to ground state [11-14]. Since the dominant recombination process is $Ce^{4+} + e^{-} \rightarrow Ce^{3+}$, the electron hole recombination may be fully suppressed. The suppression of the usual electron-hole (V-Centers) recombination results in the reduction in TL intensity [15].

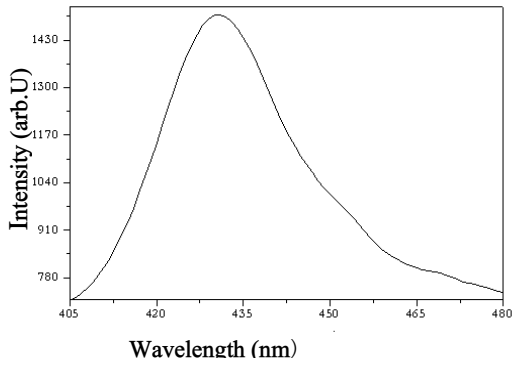

Fig. 4: TL emission curves for KI: Ce^{3+} at 445K

5. CONCLUSION

The TL glow peak at 393K is attributed to Ce^{3+} ions in the vicinity of an F-center.Ce^{3+} is considered to act as an electron trap and V-type defects (V_3 centers) as hole traps. The observed blue emission is attributed to Ce^{3+} ions. Thermo stimulated emission strongly suggesting the TL process to be the thermal release of F-electrons. Z_1-centers are formed due to F-light bleach of KI:Ce^{3+}.The TL emission under the glow peak 445K contain the characteristic blue emission of the Ce^{3+} ions, in addition to the emission due to the recombination of F electrons with their counter parts.

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