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Effect of particle size on the Lyoluminescence of γ-irradiated pure alkali halide microcrystals

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ABSTRACT : When γ-irradiated microcrystals of NaCl, NaBr, KCI, KBr and KI are dissolved in Iuminol solution, initially the LL intensity increases linearly with time, attains an optimum value, then decreases and finally disappears. The LL intensity was detected by Photo Multiplier Tube whose output was connected to x-y recorder. The LL intensity depends upon the particle size and the peak of optimum LL intensity also depends upon the particle size. Optimum LL intensity (I_m) initially increases with increasing particle size due to greater stability of colour centers in crystallites of larger dimensions. Whereas in crystallites of smaller dimension there is rapid decay of coloration. For course particle size the formation of radiolysis product is very much higher than for fine particles. Time (t_m) corresponding to the peak of the LL intensity increases with increasing particle size of alkali halide microcrystals because the rate of dissolution is very less in course particle size then in fine particle size and decay time increases with increasing grain size.

Keywords : Lyoluminescence, Luminol (3 amino-phlltalahydnazide)

INTRODUCTION

Lyoluminescence is a process in which the emission of a visible glow takes place during the dissolution of irradiated crystals in suitable solvent. It was first reported in the case of some organic crystals [1,2]. Later on enhancement of luminescence when gamma irradiated NaCl is dissolved in certain fluorescent solution [3]. The light emission takes place when a hydrated electron (e^{-}_{aq}) from the dissolving crystals recombines with a hydrated chlorine atom (Cl) which transfer its excitation energy to the fluorescent dye [4].

$$X + e^{-}_{aq} - (X_{aq})^{*} - X_{aq} + hv$$

Where X = Cl, Br, I etc.

When alkali halides are irradiated with ionizing radiation colour centres are formed in the materials it is known that the coloration produced by electrolysis is very stable [5]. It has been shown the coloration in microcrystaline power obtained by crushing the electrolytically coloured single crystal is not stable [6-10]. In case of potassium halides all the coloration was lost within a day [11]. Deshmukh and his co-worker [5-10] have established a correlation between microhardness, dislocation mobility and the stability of colour centres in microystals powder of alkali halides [12]. Chatterjee *et. al.*, [13] have a theoretical mode for the quantitative estimation of LL from organic materials.

The effect of grain size on LL from organic phosphors has been reported by Ettinger and Puttie [14]. Recent studies have been made by Balogun and Adesanmi [15] showing that dissolution stability of LL response are grain size dependent in mannose. Another factor explaining grain size dependence might be insensitive surface layers. According to the interphase model of Ahnstrom [2] which explains the mechanism of LL in NaCl, interphase reactions will take place only in limited space. Lelivere and Adloff have found that LL yield is maximum for grain size (~ 80µm) for NaCI and the decreases with further increase in grain size. The rate of dissolution has been discussed as a function of grain diameter ~50 to 400µm. Kalkar has reported that LL intensity from NaCl increases linearly with increasing mesh size in a very narrow range 50 to 200µm [17]. All the reported works on the particle size dependence LL intensity is restricted to NaCI the studies made to date are for very narrow particle size range. When luminol solution was used a great enhancement of light yield was observed which was almost five orders of magnitude greater than that produced in water. The present paper reports the effect of particle size on the LL of *Y*-irradiated NaCl, NaBr, KCl, KBr and Kl microcrystals.

EXPERIMENTAL DETAILS

The pure NaCl, NaBr, KCl, KBr and Kl microcrystals were grown from their melt using slow cooling method, *i.e.*, czochralski method. For the measurement of lyoluminescence, the crystals were crushed into microcrystals form and the microcrystals having different grain sizes were separated by using sieves of particular dimensions, then these were coloured by exposing them to ^{60}Co gamma source. To study the effect of grain size on lyoluminescence, 2 ml luminol solution was injected with the help of a syringe into the 10 mg. sample taken in a transparent glass tube placed close to the photomultiplier tube kept in a LL cell. The intensity of LL produced was detected by RCA-931 PMT whose output was connected to an *X-Y* recorder. The process was repeated for different grain sizes.

RESULTS

Fig.1 shows time dependence of the glow curve of γ-irradiated microcystals of pure NaCl for different grain sizes, respectively. When γ-irradiated microcystals are dissolved in a fixed volume of luminol solution, then initially the LL intensity increases with time, attains a maximum value, later on it decreases and finally disappears.

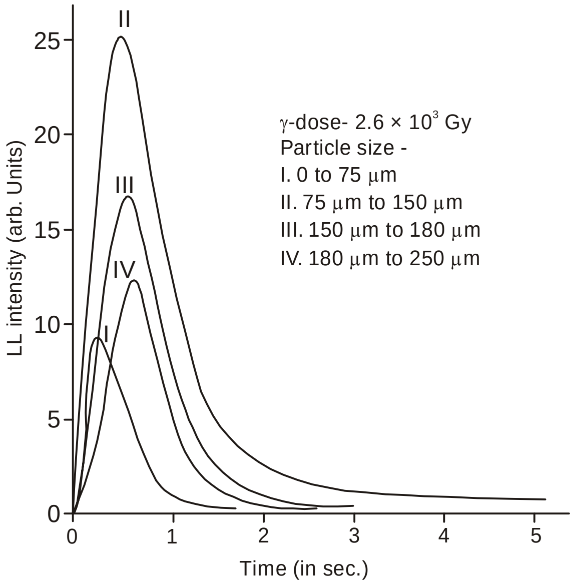

Fig.1. Time dependence of LL intensity of γ-irradiated NaCl sample for different particle size.

Fig.2 shows the dependence of LL intensity l_m versus particle size. The peak value of LL intensity l_m initially increases with increasing grain size, attains an optimum value, then it decreases with increasing grain size.

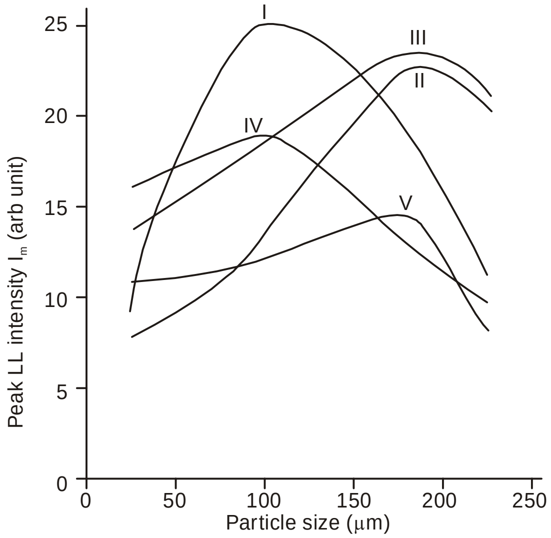

Fig.2. Variation in peak LL intensity I_m with particle size of I. NaCl, II. NaBr, III. KCl, IV. KBr, V. KI.

Fig.3 shows the dependence of time t_m are particle size. It is seen from the Fig. that the value of t_m increases with increasing grain size of microcystals of samples.

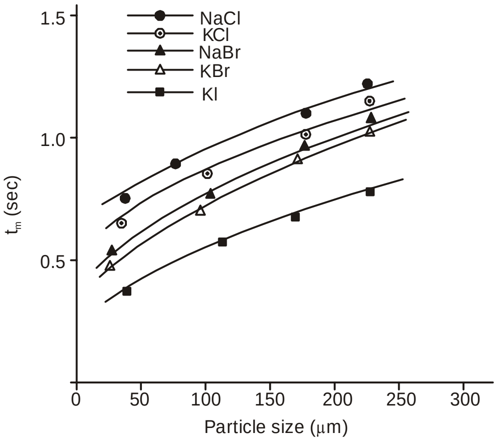

Fig.3. Dependence of t_m of pure alkali halides on the grain size.

Fig.4 shows the plot of Log-*l* versus (*t-tm*) for microcystals for different grain sizes. It is seen that the plot between log-l versus (*t-tm*) is a straight line with a negative slope. This plot indicates the exponential decay of LL intensity which can be expressed by the relation :

$$l = l_o \exp[-(t - t_m)/\tau]$$

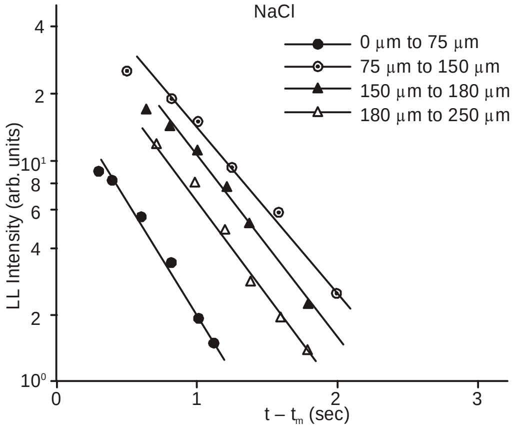

Fig.4. Plot of log I versus (t – t_m) of different grain size of NaCl microcrystals.

Fig.5 shows the dependence of decay time (τ) of LL intensity on different grain sizes of NaCl, NaBr, KCl, KBr and Kl. It is seen that ô increases with increases in particle size.

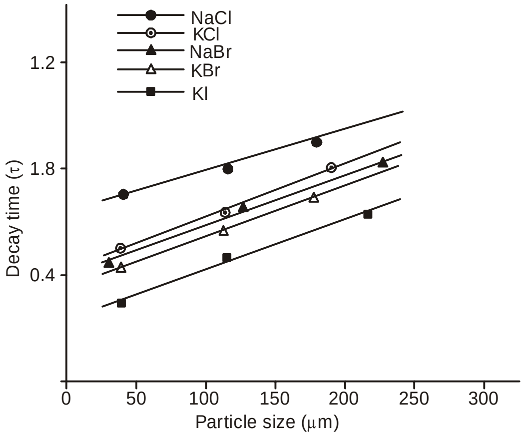

Fig.5. Dependence of decay time of LL intensity on the different size of pure alkali halide microcrystals.

DISCUSSION AND CONCLUSION

When γ-irradiated microcrystals of alkali halides were dissolved in luminol solution, the LL emission is found in which electrons are released from *F*-centres and the formation of hydrated electrons takes place. The hydrated electron radiatively recombines with the holes on the surface of the crystallites and gives rise to LL emission. The mechanism of LL may be described as :

$$F-\text{centre}\xrightarrow[\text{Recombination}]{\text{Hydration}} e_{aq}^{-}$$

$$e_{aq}^{-}+V_2-\text{centre}\xrightarrow[\text{(Water – Solid interface)}]{\text{Recombination}} h\nu$$

The expression for l, peak intensity of LL (l_m), and (l_m), are as given below :

$$l = \eta\,\gamma\,\alpha\, n_F N_o \exp\,[-(t-t_m)/\tau] \qquad \text{...(1)}$$

$$l_m = \eta\,\gamma\,\alpha\, n_F N_o \qquad \text{...(2)}$$

$$t_m = \frac{1}{(\beta-\alpha)}\ \text{In}\ (\beta/\alpha) \qquad \text{...(3)}$$

Where β is the rate constant for the recombination of hydrated electrons with holes, $\alpha = 1/\tau$, is the rate dissolution of solute in the solvent, η is the probability of radiative recombination, γ is the factor correlating the number of the hydrated electrons, n_F is the density of *F*-centres, and N_o the initial number of the molecules of solute.

Equation (1) shows that when luminol solution will be dropped into the solute then intially the intensity should increase with time, attains a maximum value and then it should decrease exponentially with time such results are shown in Fig.1.

Fig.2 shows that, initially l_m increases with increasing particle size of alkali halide. This increase in peak of LL intensity may be due to grater stability of colour centres in crystalities of larger dimensions where as in crystallities of smaller dimensions, there is a rapid decay of coloration because of bleaching of colour centres may take place. Equation (2) indicates that l_m should decrease firstly due to the formation of less density of colours centres in larger grain size and secondly due to decrease in the dissolution rate α with increasing grain size of microcrystales.

It is seen from Fig.3 that the time t_m of the LL intensity increases with increasing particle size of samples. This may be due to longer time duration needed for dissolving the sample of larger particle size. As the rate of dissolution of solute decreases with increasing particle size, Equation (3) shows that l_m should increase linearly with particle size of sample.

Fig.5 shows that decay time τ increases with grain size. Equation (1) shows decay time τ should inversely depend on rate of dissolution (α) of solute in the solvent.

It can be concluded that an enhancement in LL intensity depends on the colour centre concentration in the powder. For coarse particle size formation of radiolysis product is much higher than for finer particles. However rate of dissolution is very much less in the farmer than later.

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