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Effect of lyoluminescence decay in KCl microcrystalline powder in lyoluminescence dosimetry of ionization radiations

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Abstract The lyoluminescent intensity of gamma irradiated potassium chloride powder of different particle sizes have been investigated In the mesh range 75-250 μm, the lyoluminescence (LL) peak intensity increases with increasing particle size Above 250 μm and upto 425 μm, the LL peak intensity decreases For particle size greater than 425 μm, the LL peak intensity increases again The LL glow curves show that LL decay times consist of two components for all particle sizes The dependence of decay time, especially the longer component on the particle size has been investigated Particle size has no significant effect on the shorter decay component in LL emission A possible explanation for the experimental results has been attempted

Keywords Lyoluminescence, KCl.

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When strongly energised alkali halide crystals are dissolved in a liquid solvent (like water), light is emitted as a result of recombination of hydrated electrons with the holes on the surface of crystallites. This phenomenon is called lyoluminescence (LL) [1] Later on, Ahnstrom [2] and Arnikar *et al* [3] also proposed the mechanism of LL. Accordingly, the electrons trapped at anion vacancies in ionic crystals get hydrated on dissolution and then react with holes forming hydrated anions in an excited state which leads to the emission.

$$X + e_{aq} \rightarrow (X_{aq}^{-})^{*} \rightarrow X_{aq} + h\upsilon ,$$

where X = halogen atoms.

The LL single obtained during the dissolution of previously irradiated materials is expressed in the form of a glow curve. These curves begin with a high peak followed by an exponentially decreasing intensity. Quite a number of studies have been undertaken to understand the LL emission [4]. However, the survey of literature shows that the particle size studies have been mainly restricted to organic compounds. The effect of particle size on LL from organic phosphors has been noted by Ettinger[5]. An important factor is the fraction of energy deposited in surrounding air as compared with energy imparted to the solid Recent studies have been reported by Balogun and Adesanmi [6] showing that dissolution curve, light yield, dose response curve and stability of LL response are particle size-dependent in the case of mannose. Results showed lower yield for finer particle size as found in earlier results reported by Ettinger [5]. Another factor explaining particle size-dependence might be insensitive surface layers. According to Ahnstrom[2], surface effects and therefore particle size will be an important parameter in LL from inorganic phosphors as it was in organics. Lelievre and Adolff [7] show an optimum particle size of ~ 80 μm for NaCl and also discuss the dissolution as a function of the grain size. For the development of LL dosimetric material investigation of different parameters are required such as density of colour centres, rate of dissolution of the sample, temperature

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of solution and the mass of the dissolved sample. Out of, these dissolution rate of the sample is dependent on the particle size of the powder. Therefore in the present work, we have investigated the dependence of LL intensity on the particle size of KCl powder with water as solvent. Since decay time is an important parameter for LL dosimetry, it would be instructive to see whether LL decay time experiments with KCl powder of different particle sizes would indicate the dependence of decay time on the particle size of microcrystals.

KCl crystals were grown by slow cooling of melt. Single crystals were crushed and particles of the desired size were obtained using test sieves. The samples were exposed to γ-rays from ^{60}Co source at a dose rate of 0.2806 x 10^4 Gy/hr. For measuring LL intensity, a small quantity of coloured powder was placed in a highly transparent glass tube which was placed close to the photomultiplier tube kept in a light tight box The box had a circular hole on the top surface through which a syringe was inserted to inject the solution into the glass tube. The intensity of light emitted during dissolution was detected by the photomultiplier tube (Hmamatsu R1307) whose output was connected to an X-Y recorder. For all measurements, 3 c.c of distilled water was added to 50 mg of the sample The experiments have been carried for eight different particle sizes. All these measurements have been repeated several times in identical experimental conditions to ensure reproducibility.

Figure 1 shows the typical LL glow curves for KCl powder exposed to γ-rays of 7 x 10^3 Gy dose for different particle sizes. The Figure 1 shows that when γ-irradiated microcrystals are dissolved in a fixed volume (3 c.c.) of distilled water, initially the LL intensity increases with time, attains a maximum value I_m at a particular time t_m, then it decreases and finally disappears. Width of the LL glow peaks increases with increasing particle size of KCl powder due to rate of dissolution of microcrystalline powder in the solvent. With increasing particle size of microcrystalline powder, the LL peak intensity increases due to the formation of radiolysis products during γ-ray irradiation, but above 250 μm size of the KCl powder, the LL intensity decreases and then becomes constant. The variation of LL peak intensity in Figure 2 (curve a), shows that above 250 μm size of microcrystalline powder of KCl, the LL intensity may vary with radiolysis products as well as by the dissolution rate. Above 400 μm, LL intensity becomes approximately constant due to increasing radiolysis products and decreasing dissolution rate in water as a solvent.

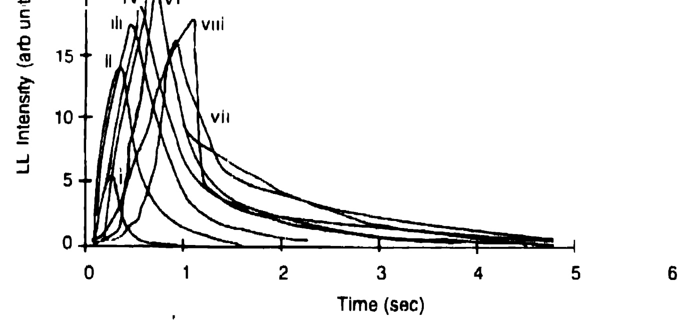

Figure 1. LL glow curves of KCl microcrystalline powder of different particle size with water used as solvent, exposed to γ-dose = 7 x 10^3 Gy Particle sizes · (i) 0-75 μm, (ii) 75-150 μm, (iii) 150-180 μm, (iv) 180-212 μm, (v) 212-250 μm, (vi) 250-300 μm, (vii) **355-425** μm and (viii) 425-850 μm

Figure 2 (curve b) shows the variation of t_m with particle size of KCl powder with water as solvent. Value of t_m increases with particle size of KCl powder. The increasing order of the curve with the particle size is due to recombination of radiolysis products which is much more in the surface of microcrystalline powder.

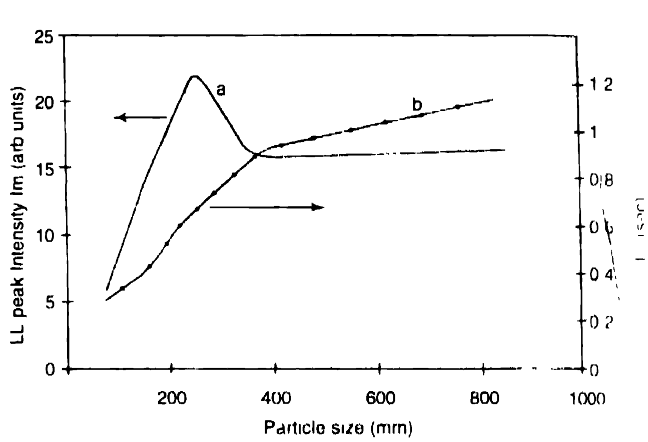

Figure 2. Variation in LL glow peak heights (I_m) (curve a) and time t_m (curve b) with particle size of KCl with water used as solvent, exposed to γ-dose = 7 x 10^3 Gy.

Figure 3 (curve a) shows that the total LL intensity (I_T) increases with particle size upto 300 μm It may be due to increasing size of microcrystalline powder being dissolved which increases the rate of recombination of radiolysis products upto 300 μm size. I_T decreases with further increase in particle size of microcrystal from 300 to 400 μm. Above 300 μm, the rate of radiolysis product increases, while rate of dissolution decreases

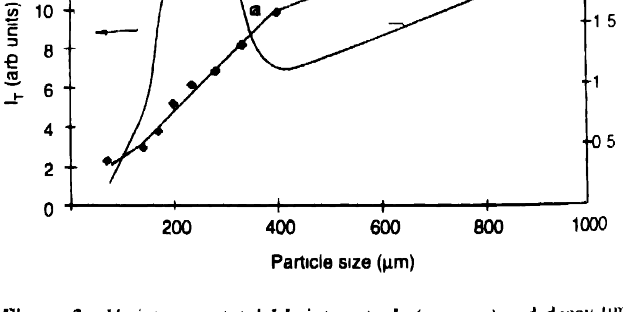

Figure 3. Variation in total LL intensity I_T (curve a) and decay time τ (curve b) with particle size of KCl with water used as solvent, exposed to γ-dose = 7 x 10^3 Gy.

Above 400 μm size of microcrystalline of KCl. I_T increases due to increase the radiolysis products during γ-irradiation, while in this region, dissolution rate decreases due to increase of particle size. The increasing rate of I_T by radiolysis products is more as compared to the decrease by dissolution rate, then final results of I_T may increase above 400 μm size of microcrystaline powder.

Figure 4 shows the plot of log I of KCl powder with time. The value of log I is increasing with particle size as more quantity of the powder is getting dissolved. The slope of the curve between log I and dissolution time is decreasing with increasing particle size, because larger sized grains dissolve slowly. This slope indicates the stability in the rate of formation of radiolysis products and dissolution of grains for the KCl powder. Therefore, decay time of LL intensity is one of the important factors for LL dosimetry.

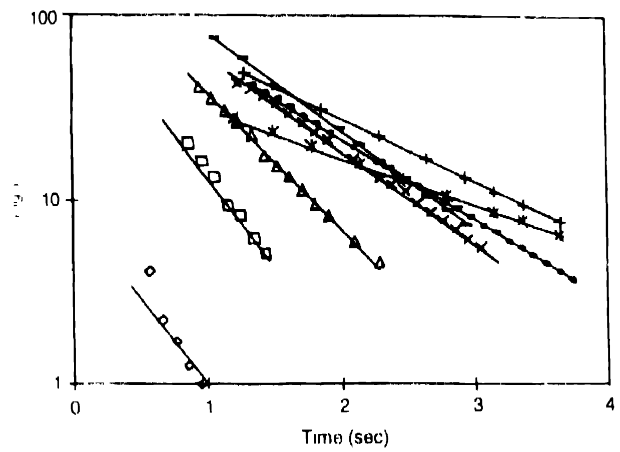

Figure 4. Plot of log I versus t (t >tm) of KCl microcrystalline powder for different particle sizes ◊ 0 – 75 μm □ 75 – 150 μm, Δ 150 – 180 μm , X 180 – 212 μm, ◆◆ 212-250 μm, – 250 300 μm , + 355 - 425 μm , * 425 850 μm

After attaining the LL peak at maximum intensity (Im), the decay of LL intensity depends on the nature of particles (Figure 3 curve b). Decay time increases upto 400 μm then saturates at higher particle size of KCl powder (above 400 μm). In Figure 2, both the curves are of increasing order with increasing size of particles due to decreasing dissolution of microcrystalline powder in water. The area between both the curves indicate the recombination of hydrated electron and hole in the surface of microcrystalline powder and recombination of radiolysis products during dissolution. This difference is very much affecting the exact energy absorption by microcrystalline powder during γ-ray irradiation, the LL intensity being not directly related to absorbed dose of powder. Therefore, fixed range of particle size (maximum variation 50 μm) may be permissible for KCl powder to be used as lyoluminescence dosimetry of ionization radiations. The LL of alkali halide crystals depends on different parameters such as radiation dose, density of colour centres, rate of dissolution, temperature of the solution, mass of the dissolved sample, type of the impurity present, pH of the solvent and type of the sample. Among them, the most important parameter is particle size of the LL phosphor.

From the results presented above, it may be concluded that the lyoluminescence intensity cannot be directly correlated to the radiolysis product (colour centre concentration) or the dissolution rate but depends on both factors simultaneously. Such studies would be helpful in providing information for lyoluminescence dosimetry and better insight into the kinetics of reactions responsible for LL emission. It would definitely add to our knowledge of defect interactions in general and particularly in solids.

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