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THINDOWN IN LITHIUM FLUORIDE (TLD-100)

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ABSTRACT

A track structure model has been applied to calculate the relative thermoluminescent efficiency of TLD- 100 irradiated with energetic heavy ions. In earlier work we proposed that the response of TLD- 100 to γ - ray arises from a combination of 1- hit and 2- hit trap structures. The supralinear (2- hit) response is now known to diminish with a decrease in the energy of incident electrons. Since the delta rays of slow particles are of low energy, diminishing with a reduction in ion speed, we calculate the response to slow heavy ions by track theory as due to the 1- hit targets alone and obtain good agreement between theory and experiment. The branching of the experimental curves is principally caused by the kinematic limit in delta ray production for ions approaching the end of their range, an effect which is called "thidown" when seen in the tracks of heavy ions approaching the end of their range in electron sensitive nuclear emulsion.

Keywords: TLD- 100 Supralinear response Track structure Relative thermoluminescent efficiency

I. INTRODUCTION

The thermoluminescent (TL) response of Lithium Fluoride to radiations with different LET's has been studied by many authors. Most of the studies, however, focus upon the variation of the supralinearity of the dose response relationship with changes in the radiation quality^[1,2]. The supralinear response of TLD- 100 (LiF) diminishes with the energy of the incident radiations. It vanishes entirely for electrons of less than 3 keV. At the same time the low dose linear response of this material remains essentially independent of the energy of incident photons or electrons. Larsson and Katz^[3] proposed earlier that the response of TLD- 100 to γ - ray could be described as due to a combination of 1- hit and 2- hit trap structures. The former requirs the passage of a single electron, while the latter requirs the passage of electron pairs. The same logical structure is used here, but we would presently add that the 1- hit response is energy independent, while the 2- hit response depends strongly on the energy spectrum of incident electrons.

In our application of track theory to the effects of stopping heavy ions, we note that delta ray energy is nominally limited kinematically to β ² MeV, where β is the ratio of the speed of the incident ion to the speed of light, while the average delta ray energy is very much lower, the number of delta electrons varying inversely with the

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square of the electron energy. For ions of energy about 5 MeV/u the limiting delta ray energy is about 10 keV, while at 10 MeV/u the limiting delta ray energy is about 22 keV. A precise formula for the energy spectrum of delta rays from energetic protons between 5 keV and 5 MeV in arbitrary gases is given by Rudd^[4].

In the present calculation we are interested in the response of TLD- 100 to stopping ions whose initial energy is below 10 MeV/u and are principally concerned with ions of lower initial energy. Because of the low average energy of delta rays from these ions we neglect the contribution to TLD response from supralinear (2- hit) traps and make our calculation as if only the linear (1- hit) traps are involved in the response of these stopping ions.

II. THE TRACK STRUCTURE THEORY IN LIF

1. Radiosensitivity parameters for TLD-100

In general, the TL yield after uniform gamma irradiation to a dose D can be represented as.

$$TL(D) = R \cdot P(C = 1, D/E_1) + (1 - R) P(C = 2, D/E_2)$$
 (1)

The first item stands for the 1- hit component having a fraction R and the second the 2- hit component. The activation probabilities P(C,D) are given from the cumulative Poisson distribution as

$$P(C=1, D) = 1 - \exp(-D/E_1)$$
 (2)

and

$$P(C=2, D) = 1 - (1 + D/E_2) \exp(-D/E_2)$$
 (3)

Examples of the best fitting values of E_1 , E_2 and R to the data of several different experiments with TLD- 100 (5,6,7,8) are given in Table 1.

 ${\bf Table~1}$ Radiosensitivity parameters for TLD-100, from the data of several authors

| Author | $E_{l}\left(Gy ight)$ | $E_{2}(Gy)$ | R |
|-----------------|------------------------|-------------|-------|
| Cameron, 1967 | 10 | 24 | 0.015 |
| Montret, 1980 | 10 | 150 | 0.008 |
| Zimmerman, 1971 | 10 | 250 | 0.003 |
| Wingate, 1967 | 10 | 300 | 0.005 |

Fig.1 shows the fit of calculations from these parameters to the data of Montret-Brugerolle. Similar fits are obtained for the other data, using the appropriate parameters. In all cases the relative contribution of 1- hit component, R, is very small. While the characteristic dose E_2 for the 2- hit component varies with the materials used by different investigators, the characteristic dose for the 1- hit component, E_1 , remains constant at 10 Gy. This is the dose of γ - ray at which there is an average of 1- hit per 1- hit target.

In track theory we make use of a second parameter, a_0 , the radius of the sensitive target. From Suntharalingam and Cameron, the concentration of the Mg^{2+} impurity in

TLD- 100 is about 100 ppm. That is the Mg²⁺ impurities are separated, on the average, by a distance of about 22 lattice constants, or about 85nm. We assume the target radius to be 40 nm, and the energy deposited within this distance can migrate to the impurity site and cause the excitation which ultimately results in the TLD response.

2. The track structure model

In a 1- hit detector for an atom of radius a_o with a radial distance t from its centre the path of an ion effective charge

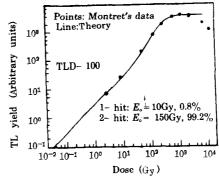


Fig.1 Parametric fit (line) to the data of Montret for the response of TLD-100 to γ -ray

 Z^* and speed β c, the probability of activation is

$$P(t) = 1 - \exp \left[- E(Z^*, \cdot, \cdot, t, a_0) \right] / E_1$$
 (4)

The action cross section, σ , can be calculated by integrating P(t) over the entire range of the delta-rays,

$$\sigma = \int_{t \min}^{t \max} 2\pi \ t P(t) dt$$
 (5)

Where t_{max} is the maximum range of the delta-rays in the medium and $t_{\text{min}} = 10^{-12} \text{m}$ in this work.

The probability that an action is generated by the beam in a 1- hit detector is

$$P = i - \exp(-\sigma F) = 1 - \exp(-\sigma D/L)$$
 (6)

where F is the ion fluence and L the stopping power, and D = FL.

The radiosensitivity, K, of the 1- hit trap structure of the detector to γ - rays is

$$K_{r} = 1/E_{1} \tag{7}$$

and the radiosensitivity to heavy ions is

$$K_{i} = \sigma / L \tag{8}$$

We define the relative TL efficiency as,

$$RTLE = K_1/K_7 = \sigma E_1/L \tag{9}$$

3. Radial dose distribution in LiF

In the calculations of the cross-section, a new formula was used to describe the dose distribution in LiF. It was based on an expression for the radial dose distribution in water, with appropriate corrections against the atomic number and density of the medium. The new expression is normalized to yield the stopping power of protons in LiF to within 10%.

Fig.2 shows the radial distribution of average dose E in a short cylindrical target of radius 40 nm. Its axis is parallel to the proton path with a distance of t. The protons travel at $\beta = 0.05$, 0.1 and 0.5, in Lithium Fluoride (TLD- 100). E for H, He,

Ne and Kr ions at $\beta = 0.1$ in TLD- 100 is shown in Fig.3. These are the typical dose distributions to obtain, by applying Eqs.4 and 5, the cross sections for track segment bombardment.

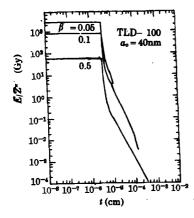


Fig.2 Calculated values of the average dose E divided by the square of the effective charge, in a target cylinder of radius 40nm whose axis is parallel to the ion's path, for ions of effective charge Z^* passing through LiF at relative speeds of $\beta=0.05$, 0.1 and 0.5, as a function of the radial distance of the axis of the cylinder from the ion's path

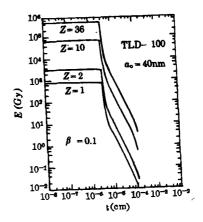


Fig.3 See caption to Fig.2. E vs. t for ions of atomic number 1, 2, 10, 36 moving at relative speed $\beta=0.1$. If a horizontal line is drawn at 10 Gy one can approximate a track "width" for each ion from the intersection of this line and the plotted curves.

4. Relative thermoluminescent efficiency

In general, the thermoluminescent efficiency, RTLE, of heavy ions in TLD- 100 is calculated from Eq.9 for thin detectors.

Montret- Brugerolle evaluated^[6] the relative TL efficiencies of He, Ne and Kr ions in thick detectors by comparing the light yields of the heavy ions to that of γ - ray at low doses (below the supralinearity region which occurs above 10 Gy for TLD- 100). In our approach, we average the response over the path length to find

$$\vec{\sigma} R = \int_0^R \sigma dr = \int_{T_i}^0 \sigma / L dT$$
 (10)

where Ti, the initial kinetic energy of the ions, and R, the ranges.

We represent the average radiosensitivity K, for stopping particles as,

$$\overline{K}_{i} = \overline{\sigma} R/T_{i} = \overline{\sigma} /\overline{L}$$
 (11)

where we implicitly define average value of LET as

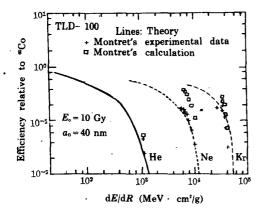
$$\overline{L} = T/R \tag{12}$$

Thus, the relative TL efficiency can be calculated as

$$RTLE = \overline{\sigma} E_1/\overline{L}$$
 (13)

III. RESULTS AND DISCUSSION

In Fig.4 we compare our calculated values of RTLE, shown as interrupted lines, to the measured values of Montret. The agreement of theory with experiment is gratifying, and lends some support to the concept of 1- hit and 2- hit targets as responsible for the supralinear response of TLD- 100 to γ - ray. In that figure we also show the calculated values of Fain, Montret and Sahraoui⁽¹⁰⁾ in which no separation was made of the 1- hit and 2- hit response characteristics. Instead, they applied the entire dose- response function after gamma irradiation to their calculation of the radial dose distribution, without taking target size into account. Their calculated efficiencies differ from the experimental findings by as much as factor of 2.



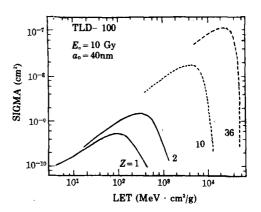


Fig.4 The relative response of TLD-100 to Fig.5 stopping He, Ne, and Kr ions as measured by Montret-Brugerolle (+) as calculated by Fain, Montret, and Sahraoui (_), (called Montret's calculation), and as calculated in the present work (interrupted lines)

The calculated action cross section vs LET for ions of atomic number 1, 2, 10, and 36 in TLD-100. The "hooks" at 'high LET arise from thindown. They do not arise from the Bragg' peak in energy deposition

In Fig.5 we show our calculated cross sections of four ions species in TLD- 100. These action cross all exceed the cross sectional area of the target (5×10⁻¹¹ cm²), so all cross sections lie in the track width regime. This is consistent with the concept of thindown. Only when tracks lie in the track width regime, when the appearance of the track resembles a "hairy rope" rather than a string of randomly placed beads (the grain count regime), it is possible for the cross section to diminish with an increase in LET. We see the effect here as a hook shaped plot of cross section vs LET, in the high LET region of the plotted curves, at the right. Similar effects have been observed for NaI(Tl) scintillators^[11], biological cells^[12], and nuclear emulsions^[13]. A theoretical analysis of particle tracks in nuclear emulsions was given by Katz and Kobetich^[14].

A plot of cross sections vs LET displays the same branching with the atomic number of the bombarding ion as a plot of relative efficiency vs LET as defined in Eq.13. When σ is plotted against the relative speed β the action cross-sections decrease in a systematic way with a decrease in β , and there is no branching with Z in the limit of low β , as shown in Fig.6. Here we see clearly that the effect is the one that depends on the ion speed rather than its LET. The difficulty raised by the plots of relative efficiency vs LET arise from the a-priori assumption that TLD effects arise from energy deposition along the ion's path. Here we see that such an assumption fails. The effects observed are largely due to delta rays, and are best interpreted through the concept of an action cross section rather than through the energy deposition in small target volumes along the ion's path.

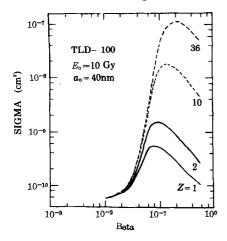


Fig.6 Calculated action cross sections vs. β for ions of atomic number 1,2,10,36

The confusing hooks observed in LET plots at high LET (low ion speeds) disappear when the cross section is plotted against β . Since at the same ion speed the LET's vary as Z^{\star} , the curves for different ions are displaced from each other when cross sections or relative response is plotted against LET.

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REFERENCES

- [1] N.Suntharalingam and J.R.Cameron, Phys. Med. Biol., 14 (1969), 397-410.
- [2] J.B.Lasky and P.R.Moran, J. Appl. Phys., 50 (1979), 4951-4957.
- [3] L.Larsson and R. Katz, Nucl. Instr. Meth., 138 (1976), 631-636.
- [4] M.E.Rudd, Phys. Rev., A, 1989, in press.
- [5] J.R.Cameron et al., Proc. int. conf. luminescence dosimetry, USAEC Conf- 650637, 1967, p.47- 56.
- [6] M.Montret-Brugerolle, Distribution spatiale de L'energie deposee par des ions energetiques dans les milieux, condenses Etude par thermoluminescence, Universite de Clermont-Ferrand II, 63170 Aubiere, France, 1980.
- [7] J.Zimmerman, J. Phys. C: Solid State Physics, 4 (1971), 3277- 3291.
- [8] C.L.Wingate et al., Proc. int. conf. on luminescence dosimetry, USAEC Conf- 650637, 1967, p.421-434.
- [9] R.Katz et al., "An analytic representation of the radial distribution of dose from energetic heavy ions in Water, Si, LiF, and NaI," Radiation Physics and Chemistry, submitted 1988.
- [10] J.Fain et al., Nucl. Instr. Meth., 175 (1980), 37-39.
- [11] E.Newman and F.E.Steigert., Phys. Rev., 122 (1960), 1575-1578.
- [12] R.Katz et al., Radiat. Prot. Dosimetry, 13 (1985), 281-284.
- [13] C.F.Powell et al., The study of elementary particles by the photographic method, Pergamon Press, New York, 1959.
- [14] R.Katz and E.J.Kobetich, Phys. Rev., 186 (1969), 344-351.