TRACK STRUCTURE CALCULATION OF THE THERMOLUMINESCENT YIELDS OF HEAVY CHARGED PARTICLES*

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Extensive effort is currently being invested in the investigation of the use of LiF thermoluminescent dosimeters (TLD) in exotic radiation fields, such as mixed neutron-gamma and high LET particulate radiation fields. For this reason it is important to investigate the relative TL response, η, to various radiation fields (η is defined as the TL signal/imparted energy by the radiation field in question/TL signal/imparted energy by Co-60 γ rays, both at low absorbed doses of the irradiated mass). The question of the universality of the TL-LET behaviour is especially pertinent to the use of LiF-TLD's in mixed n-γ or charged particle radiation fields. For example, large discrepancies exist among published fast neutron sensitivities and high energy electron sensitivities even in the cases when the problem connected with the determination of the imparted energy are claimed to be solved. Unfortunately, the possibility of non-universality of the TL-LET response has been generally overlooked, perhaps, because it has been proposed (Tanaka et al., (1) and many others) that, indeed, the relative TL-LET response curve is universal, i.e., the stopping power, S, of the directly ionizing radiation is the dominant factor that influences the relative TL response and that there is a unique curve η = η(S) common to many TLD materials. Our purpose is to show that the situation is far more complicated than the simplistic picture proposed by Tanaka et al., and others.

In the first instance many groups have reported data that illustrate that the η of a particular type of radiation depends on the type of dosimeter (e.g., LiF, $Li_2B_4O_7$, BeO, CaF_2, quartz, etc...). Furthermore, we have carried out extensive studies (2,3) irradiating various batches of LiF and $Li_2B_4O_7$ (Harshaw TLD bulb dosimeters) with 13.54 meV neutrons and 3.8 MeV alphas, which showed that purchase of a particular type of TLD even from the same supplier and with the same nominal dopant concentrations (Mg, Ti in LiF, Mn in $Li_2B_4O_7$) does not guarantee identical or even similar TL-LET response characteristics (Table 1).

Another question in the application of the dependence of η on LET of a particular dosimeter is whether the LET of the ionizing radiation is the only parameter of the radiation field that influences η, or whether other parameters of the radiation field (e.g., the velocity of the directly ionizing particles) have to be taken into account. We have therefore studied the TL induced in LiF-TLD's by fission fragments using a 5 μm mylar degraded flux from a Cf-252

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source in vacuum (4), Approximately 1/3 of the imparted energy arises from fission fragments with average specific $\overline{LET}$ in LiF-TLD of approximately $3 \cdot 10^4$ MeV $g^{-1}cm^2$. Most of the remainder of the imparted energy arises from α particles emitted from Cf-252.

Table 1. Relative Response of LiF and $Li_2B_4O_7$ to neutrons and alphas

13.54 meV Neutrons			3.8 MeV α particles		
Type	Batch	$\bar{\eta}$	Type	Batch	$\bar{\eta}$
LiF	1-TLD-100	0.340±0.007	LiF	1-TLD-100	0.170±0.024
LiF	2-TLD-600	0.420±0.007	LiF	2-TLD-600	0.210±0.007
$Li_2B_4O_7$	1-TLD-800	0.775±0.007	LiF	3-TLD-700	0.290±0.013
$Li_2B_4O_7$	2-TLD-800	1.105±0.017			

All the models predicting the dependence of η on $\overline{LET}$ and the experimental data on η versus LET indicate, to the best of our knowledge, a decrease of η with increase of LET in the region of high $\overline{LET}$. For example, the one and two trap model (5) used by Jähnert predicts relative TL efficiencies (fission fragments to 5.4 MeV alpha particles) in LiF-TLD's of 0.05 and 0.23 respectively. We have measured a value of η_{ff}/η_α = 1.26 ± 0.28 in serious contradiction to these predictions (the statistical error comprises only a small part of the indicated total error of η_{ff}/η_α.). This result together with other published experimental data hint at the multiplicity of the TL-LET function, i.e., the type of the directly ionizing particle must be taken into account in the study of η versus LET and that at least in the high $\overline{LET}$ region η increases with particle mass for a particular $\overline{LET}$.

THEORY

The reduced relative response of LiF-TLD to high LET radiations is widely explained to arise from the recombination of the liberated charge carriers by the primary heavy charged particle, or from saturation of the activator sites (which depends on the concentration of locally available activators) or from both effects. The influence of these effects on η must be treated in a three dimensional model which takes into account the delta rays, which escape from the heavily irradiated column around the path of the primary particle and produce TL signal with high efficiency. The radial distribution of absorbed dose imparted by the ejected electrons around the path of the heavy charged particle determines the track structure and is believed to be a relevant parameter to describe the radiation end effect. In the theory of track structure (6) the dose-response function of a system to gamma rays is coupled with the spatial distribution of dose from secondary electrons to yield the response of the detector to the heavy ion. Information about the ability to produce TL signal of the heavily irradiated volumes around the track can be arrived at using electron dose-TL response.

Such calculations require the knowledge of the dose-TL response dependence of the particular dosimeter under study. We define a dose response function, f(D), to be the ratio of the TL response (TL signal/imparted energy) at a particular absorbed dose from a radiation field to the TL response at low dose from the same radiation field. Since f(D) is quality dependent and the mean energy expended per ion pair formed is energy dependent for low energy electrons, the dose-TL response must be generated by electrons of initial spectrum (as far as this is possible) similar to that of the initial electrons ejected by the heavy charged particles, HCP, and not by γ rays as is usually applied in the theory of track structure (6,7). Since the maximum energy of ejected free electrons by a 4 MeV α particle is about 2 keV, it is therefore improper to generate and use a f(D) function using electrons with initial energies two orders of magnitude greater.

If the heavy irradiation in the wake of the densely ionizing HCP is the only effect that governs the dependence of η on the $\overline{LET}$ of the radiation and there exists cylindrical symmetry on the radial distribution of the absorbed dose, $D(r,\ell,W)$, imparted by the ejected particles around the track and along the path of the HCP (with average path length R in the TLD) the η of the radiation will be given by

$$\frac{\eta_\alpha}{\eta_e} = \frac{\int_0^R \int_0^\infty f(D)D(r,\ell,W)2\pi r dr d\ell}{\int_0^R \int_0^\infty D(r,\ell,W)2\pi r dr d\ell} \quad (1)$$

The radial distribution of absorbed dose in nanometric scale for various HCP (protons, α particles, I-127 ions etc...) D(r,W) have been calculated by W. Baum and collaborators (8) based on their measurements of the ionization current produced by a monoenergetic beam of particles within a small movable ionization chamber of transparent mesh located in a large cylinder filled with tissue equivalent gas at variable low pressure.

The radial distance in LiF-TLD from the data in tissue can be simulated using the relation

$$r_{LiF} = r_t \left[\frac{dE}{dx}^{e,t} \Big/ \frac{dE}{dx}^{e,LiF} \right] \quad (2)$$

where $(dE/dx)^{e,t}/(dE/dx)^{e,LiF}$ is the average ratio of the stopping power of the ejected electrons in the two media, and the radial distribution of absorbed energy in LiF, $D_{LiF}(r_{LiF},W)$ by

$$D_{LiF}(r_{LiF},W) = \left[\frac{dE}{d\xi}^{HCP,LiF} \Big/ \frac{dE}{d\xi}^{HCP,t} \right] \cdot \left[\frac{\rho_{LiF}}{\rho_{tis}}^{2} \cdot \frac{dE}{d\xi}^{e,LiF} \Big/ \frac{dE}{d\xi}^{e,t} \right] \cdot D_t(r_t,W)$$

where $(dE/d\xi)^{HCP,LiF}/(dE/d\xi)^{HCP,t}$ is the ratio of the mass stopping power of the heavy charged particle at energy W in LiF and tissue and ρ_{LiF}, ρ_t are the densities of the two materials.

RESULTS AND DISCUSSION

Preliminary results were obtained via H-3 irradiation of LiF (TLD-100). The mean energy of the emitted beta particles after appropriate backscattering corrections was calculated to be 6 keV. Beta particles were preferred over ultra-soft X-rays and lower energy electrons in order to assure that similar volumes of the TLD's were irradiated by the electrons and the HCP employed.

The calculations for 3.8 MeV alpha particles stopping in the TLD lead to values of η about two to three times greater than those we measured with the same TLD's (even allowing for a factor of two error in the dose determination, η changes by only 20%). In addition we have calculated the ratio of the TL response of 33 MeV I-127 ions to the TL response of 3 MeV α particles to be approximately 0.25, while we have found experimentally that the ratio of the TL response of degraded Cf-252 fission fragments (with mean energy of the light and heavy fragments 49 and 31 MeV respectively (4)) to the TL response of 5.4 MeV α particles (both stopping in the TLD) is five times greater. Unfortunately radial dose distribution data for I-127 ions at lower energies are not available, so that the calculation could not be carried out over the entire range of the fragments. These results were obtained with dosimeters annealed in air and there exists the possibility of surface contamination altering the TL response to HCP's and to beta particles. We are therefore repeating these experiments using more sophisticated techniques to eliminate such a possibility (10).

Tochilin et al., (9) measured the dose response function of BeO to X-rays with effective energy $\simeq$ 9 keV and found for 10.4 MeV/amu C-12 and Ne-20 ions $\eta = 2.06 \pm 0.14$ and 2.25 ± 0.38 respectively. Application of our method using data for 9 MeV/amu O-16 ions leads to $\eta = 1.5$.

These results indicate that the main (if not the only) effect that governs the values of η is the local concentration of the imparted energy around the track of the HCP and point to the possibility of calculating the TL response of any dosimeter to any ionizing radiation field (e.g., epithermal and fast neutrons) by measuring only the dose response function to electrons. Further investigations we are carrying out in our laboratory will explore the extent and accuracy of the applicability of Track Structure Theory to thermoluminescence.

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