THERMOLUMINESCENCE SOLID-STATE NANODOSIMETRY— THE PEAK 5a/5 DOSEMETER

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The shape of composite peak 5 in the glow curve of LiF:Mg,Ti (TLD-100) following $^{90}\text{Sr}/^{90}\text{Y}$ beta irradiation, previously demonstrated to be dependent on the cooling rate used in the 400°C pre-irradiation anneal, is shown to be dependent on ionisation density in both naturally cooled and slow-cooled samples. Following heavy-charged particle high-ionisation density (HID) irradiation, the temperature of composite peak 5 decreases by $\sim 5^\circ\text{C}$ and the peak becomes broader. This behaviour is attributed to an increase in the relative intensity of peak 5a (a low-temperature satellite of peak 5). The relative intensity of peak 5a is estimated using a computerised glow curve deconvolution code based on first-order kinetics. The analysis uses kinetic parameters for peaks 4 and 5 determined from ancillary measurements resulting in nearly ‘single-glow peak’ curves for both the peaks. In the slow-cooled samples, owing to the increased relative intensity of peak 5a compared with the naturally cooled samples, the precision of the measurement of the 5a/5 intensity ratio is found to be $\sim 15\%$ (1 SD) compared with $\sim 25\%$ for the naturally cooled samples. The ratio of peak 5a/5 in the slow-cooled samples is found to increase systematically and gradually through a variety of radiation fields from a minimum value of 0.13 ± 0.02 for $^{90}\text{Sr}/^{90}\text{Y}$ low-ionisation density irradiations to a maximum value of ~ 0.8 for 20 MeV Cu and I ion HID irradiations. Irradiation by low-energy electrons of energy 0.1–1.5 keV results in values between 1.27 and 0.95, respectively. The increasing values of the ratio of peak 5a/5 with increasing ionisation density demonstrate the viability of the concept of the peak 5a/5 nanodosimeter and its potential in the measurement of average ionisation density in a ‘nanoscopic’ mass containing the trapping centre/luminescent centre spatially correlated molecule giving rise to composite peak 5.

INTRODUCTION

Ionisation density-dependent phenomena in the thermoluminescence (TL) characteristics of LiF:Mg,Ti (TLD-100) are well known^(1, 2). The most intensively investigated include: (i) the linear/supralinear dose response following photon/electron irradiation; (ii) the decrease of the dose response supralinearity (decreasing TL efficiency) with decreasing electron/photon energy below ~ 100 keV, (iii) the decreasing heavy-charged particle (HCP) relative efficiency at high-ionisation density (HID) and (iv) the ‘over-response’ in the TL efficiency with decreasing X-ray energy appearing again below ~ 100 keV. All of these phenomena have been successfully modelled using microdosimetric theory^(1, 2) in the framework of the Unified Interaction Model (UNIM), Track Structure Theory (TST) and microdosimetric ‘target’ theory, respectively. Recently, however, some reservations have been voiced concerning the basic premise of TST which ignores the effects of nuclear scattering on the relative HCP TL efficiency⁽³⁾. The linear/supralinear behaviour of the dose response and its dependence on ionisation

density have been interpreted in the UNIM as owing to increased localised recombination with increasing ionisation density, an explanation which is also effective in describing the equally subtle phenomenon investigated in this paper, i.e. the effect of ionisation density on the shape of composite peak 5 (specifically the ratio of the intensity of peak 5a to peak 5). It is suggested that this ratio has the potential of serving as a measure of average ionisation density in a ‘nanoscopic’ mass containing the trapping centre (TC)/luminescent centre (LC) spatially correlated molecule giving rise to composite peak 5. Composite peak 5 (believed to arise from a TC/LC configuration based on $\text{Mg}^{++}\text{Li}_{\text{vac}}$ trimers (the TC) coupled to $\text{Ti}(\text{OH})_n$ (the LC)⁽²⁾ consists of the main peak 5, a low-temperature satellite (peak 5a) and a high-temperature satellite (peak 5b). Following low-ionisation density (LID) irradiation, peak 5 appears at a temperature of maximum intensity, T_{max} , of $\sim 208^\circ\text{C}$ when the sample is naturally cooled following the 400°C pre-irradiation anneal and then heated at a linear heating rate of 1°C s^{-1} . Peaks 5a and 5b appear at values of T_{max} of

$\sim 196^\circ\text{C}$ and $\sim 224^\circ\text{C}$, respectively. The relative intensity of peak 5a induced by (LID) $^{90}\text{Sr}/^{90}\text{Y}$ radiation in naturally cooled materials is $\sim 0.04 \pm 0.02$ (1 SD) and has proven difficult to measure reproducibly even with carefully controlled annealing, readout and deconvolution protocols because of its low intensity and strong overlap with peaks 4 and 5. Following HID irradiation, the relative intensity increases to values of 0.12 and 0.37 for 7.5 and 2.0 MeV alpha particles, respectively. Peak 5b is readily observed only at high LID dose levels or following HCP irradiation. Typical deconvoluted glow curves for naturally cooled materials and following LID and HID irradiations are shown in Figure 1. By natural cooling, is usually meant the removal of the samples from the annealing oven at the end of the pre-irradiation anneal. The sample temperature decreases

approximately exponentially and the cooling rate will be mainly determined by the mass and heat capacity of the sample holder. The average cooling rate is $\sim 1000^\circ\text{C h}^{-1}$. As previously mentioned, the effect of ionisation density on the shape of composite peak 5 is quite subtle and easily overlooked and/or ignored⁽⁴⁻⁶⁾. For low-energy alpha particles ($\sim 1 \text{ MeV amu}^{-1}$), the T_{max} of composite peak 5 is shifted to lower temperatures by $\sim 5^\circ\text{C}$ and the width of the peak is increased. The shoulder on the low-temperature side of peak 5 (owing to peak 4) is no longer observed. This behaviour can be interpreted as due to an increased intensity of peak 5a relative to peaks 4 and 5 following HID irradiation. The deconvolution analysis into component glow peaks shown in Figure 1 is based on this hypothesis and is supported by $T_{\text{m}} - T_{\text{stop}}$ studies⁽⁷⁾, which unambiguously show an additional peak between peaks 4 and 5 following HID irradiation.

Figure 1. Deconvoluted glow curve of LiF:Mg,Ti (TLD-100) into component glow peaks following irradiation by $^{90}\text{Sr}/^{90}\text{Y}$ beta (top) and alpha particles (bottom) in 'naturally cooled' samples. Note the enhanced intensity of peak 5a following the alpha particle irradiation.

THE PEAK 5A IONISATION DENSITY-DEPENDENT NANODOSEMETER

It has been previously demonstrated that peak 5a arises from geminate recombination between a locally trapped electron and hole (a 'two-energy transfer' event in microdosimetric terminology⁽⁸⁾) in a TC/LC complex of $\sim 2 \text{ nm}$ dimensions⁽⁹⁾. Peak 5 on the other hand results from delocalised recombination arising from a singly trapped electron in the TC/LC complex (a 'single-energy transfer' event) and peak 4 from a 'singly trapped hole' in the same complex (Figure 2). Localised recombination via tunnelling is a well-known phenomenon in luminescence mechanisms and is usually described in the terminology of 'donor-acceptor pairs' in which the probability of recombination is strongly dependent on the distance between the donor and the acceptor⁽¹⁰⁾. In the geminate recombination mechanism responsible for peak 5a, delocalised charge carrier traffic in the conduction band/valence band is not involved, whereas the opposite is true for peaks 4 and 5 in which the recombination mechanism is completely delocalised. The TC giving rise to composite peak 5 is associated with a 4 eV optical absorption band and very strong conversion of peak 5a to peak 4 following 4 eV optical excitation and stepped ($I_{\text{m}} - T_{\text{stop}}$) annealing is the primary experimental evidence supporting the above hypothesis for the TL absorption mechanisms giving rise to composite peak 5⁽¹¹⁾. Owing to the 'two-energy transfer event' nature of peak 5a, it is expected on purely statistical grounds that its relative intensity following HID irradiation would be increased relative to the same ratio following LID radiation because of the very high levels of dose (ionisation density) in the HCP track⁽¹²⁾. Model calculations of the ratio of peak 5a/5 following

Configuration states after irradiation

Figure 2. Schematic representation of the molecular TC/LC complex giving rise to peak 4 and composite peak 5. Following irradiation, the complex can capture an e-h (giving rise to peak 5a)—an electron-only (peak 5), a hole-only (peak 4) or be un-occupied. The linear dose response is due to e-h recombination in the e-h-occupied complex. Reproduced from Horowitz and Olko⁽¹⁾.

proton and alpha particle irradiation using a track structure approach have been carried out demonstrating enhanced intensity of peak 5a following HCP irradiation owing to full population of TCs and LCs and de-activation of CCs at HID⁽¹³⁾. Very high values of the ratio of 5a/5 of ~ 0.5 following 100 keV X-ray irradiation at dose levels > 5000 Gy have also been experimentally observed in naturally cooled material⁽¹⁴⁾. The relative intensity of peak 5a to peak 5 thus represents a measure of ionisation density in a volume of nanometre dimensions (corresponding to the approximate size of the TC/LC complex), which may be correlatable with radiation damage in certain biological systems owing to its conceptual similarity to the radiation-induced mechanisms of 'single-strand breaks' and 'double-strand breaks' in DNA⁽⁸⁾. Realisation of the potential of this 'solid-state nanodosimeter' has, however, previously been hampered by the low intensity of peak 5a relative to peak 5 of 0.04 ± 0.02 (1 SD) following LID irradiation in samples that are naturally cooled following the 400°C pre-irradiation anneal⁽⁹⁾.

THE EFFECT OF SLOW COOLING

The properties of a modified LiF:Mg,Ti (TLD-100) material in which the relative intensity of peak 5a is significantly increased have been recently described^(15, 16). The sample is prepared using a slow/linear/programmer-controlled cooling rate of 100°C h⁻¹ to room temperature following the 400°C/1 h pre-irradiation anneal. A post-irradiation anneal of 155°C/6 s on the reader planchet is also applied to both simplify and improve the robustness of the deconvolution procedure. Slow cooling decreases the intensity of peak 5 and increases the relative intensity of peak 5a resulting in increased values of the ratio of 5a/5. This behaviour arises as different rates of cooling ensure different equilibrium concentrations of dipoles ($\text{Mg}^{++}\text{Li}_{\text{vac}}$), dimers, trimers, clusters and precipitates⁽¹⁷⁾. The broadening of composite peak 5 at slow cooling rates was previously reported by Bos *et al.*⁽¹⁸⁾ and Vana and Skrobanek⁽¹⁹⁾. The former carried out a systematic study on the effect of cooling rate and reported an activation energy of 1.95 eV at a cooling rate of 1000°C h⁻¹ decreasing to 1.8 eV at 60°C h⁻¹. Bos

The effect of cooling rate

Figure 3. Comparison of glow curves following 6 MeV photon irradiation in 'naturally cooled' and 'slow-cooled' samples. Note the shift of $T_{\max}$ to lower temperatures in the slow-cooled samples and the disappearance of the shoulder arising from peak 4. This is attributed to an increased intensity of peak 5a in the slow-cooled samples.

et al.⁽¹⁸⁾ suggested that the broadening of peak 5 was because of increased Mg precipitate phases bringing about a perturbation in the crystal potential and a distortion of the band gap in the vicinity of the precipitate particles. This hypothesis, although plausible, has yet to be supported by ancillary experimental evidence. The effect of slow/linear cooling rate on the glow curve following LID irradiation is shown in Figure 3. Note the shift in $T_{\max}$ of composite peak 5 to lower temperatures and the lack of a low-temperature shoulder in the slow-cooled glow curve, reminiscent of the behaviour following HID irradiation in naturally cooled material and again, indicative of an increased presence of peak 5a. Slow cooling and improved deconvolution procedures have significantly increased the precision of measurement of the ratio of peak 5a/5 as will be described in the following.

EXPERIMENTAL DETAILS

Four samples of LiF:Mg,Ti (TLD-100) 3 mm × 3 mm × 0.89 mm chips (Batch S-4453) were irradiated in a variety radiation fields. The batch sensitivity was measured by irradiation of 10 chips with ^{137}Cs gamma rays (661 keV) at the Israeli Secondary Standards Dosimetry Laboratory (ISSDL) at the Soreq Nuclear Research Center. The ISSDL

dosimetry has been calibrated via the Primary Standards Dosimetry Laboratory at the Physikalisch-Technische Bundesanstalt and undergoes annual postal intercomparisons at the International Atomic Energy Agency. The standard deviation (SD) of the composite peak 5-sensitivity for the 10 chips was measured to be 6 % and this was assumed to be representative of the entire batch. Glow curve readout was carried out on a Harshaw 3500 manual reader at a heating rate of 1°C s^{-1} from an initial temperature of 50°C to a maximum temperature of 375°C . After irradiation, each TLD was post-irradiation-annealed at $155^\circ\text{C}/6\text{ s}$ to remove the low-temperature component of the glow curves thereby simplifying the deconvolution procedure. The temperature stability of the readout procedure has been measured to be $\pm 1.75^\circ\text{C}$ (± 1 channel) by repeated measurements of the maximum temperature of composite peak 5 following $^{90}\text{Sr}/^{90}\text{Y}$ irradiation. The chips were pre-irradiation-annealed in air at 400°C for 1 h followed by natural cooling, i.e. a rapid non-linear cool-down in ~ 20 min to room temperature, achieved by removing the TLDs from the annealing oven, or by slow cooling in the oven to room temperature with a constant cooling rate of 100°C h^{-1} determined by a programmer/controller. The readout procedure was carried out in a flow of high-purity N_2 gas following passage through a dehydrator.

GLOW CURVE ANALYSIS

The relative intensity of glow peak 5a was determined by computerised glow curve deconvolution using peak-shape parameters (Table 1) based on first-order kinetics⁽²⁰⁾ and deduced from ancillary studies using 4 and 5 eV optical excitations⁽²¹⁾, which create nearly ‘single-peak’ glow curves for peaks 4 and 5. Optical bleaching at 4 eV leads to an isolated peak 4 in the glow curve, and post-irradiation thermal annealing at 400°C followed by 5 eV (F band) optical excitation leads to an isolated peak 5. A subtraction procedure then allowed the determination of the peak-shape parameters of peak 5a. These studies resulted in activation energies of 1.9, 2.3 and 2.15 eV for peaks 4, 5a and 5, respectively⁽²¹⁾. The uncertainty in these values is estimated at ± 0.1 eV for peaks 4 and 5 and ± 0.2 eV for peak 5a. The activation energies of 1.9 and 2.15 eV for peaks 4 and 5, respectively, are in good agreement with the range of values reported in the literature⁽²⁰⁾. Satisfactory fits to the glow curves in both naturally cooled and slow-cooled materials were obtained using these activation energies that were kept constant for all the analyses. The relative intensity of peak 5a is not very sensitive to the value of the activation energy of peak 5a, e.g. using a value of 2.15 eV instead of 2.3 eV changes the 5a/5 ratio following HCP irradiation by $\sim 10\%$. The values of $T_{\max}$ were allowed to vary but only within the $\pm 2^\circ\text{C}$ corresponding to the temperature stability/reproducibility of the readout procedure. This point deserves emphasis. If the value of $T_{\max}$ of peak 5 used for the analysis of the HCP-induced glow curves is allowed to vary freely, the chi-squared minimisation procedure will invariably converge on a value of $T_{\max}$ of peak 5 corresponding to the experimentally measured $T_{\max}$ of composite peak 5. This will then invariably lead to a loss in the correlation between peak 5a and ionisation density, as was reported by Brandan *et al.*⁽²²⁾, in the various

deconvolution protocols employed in that investigation where ‘the curves were displaced in order to overlap the positions of their peak 5’. The deconvolution procedure used herein is based on the hypothesis that peak 5 does not shift in temperature as a function of ionisation density and that the experimentally observed shift in the $T_{\max}$ of composite peak 5 is owing to the increased relative intensity of peak 5a, which ‘pulls’ the $T_{\max}$ of composite peak 5 downward in temperature. This procedure reveals a robust correlation of the intensity of peak 5a with ionisation density and has been successful in leading to improved precision of measurement of the 5a/5 ratio in both types of materials and was found to produce satisfactory fits to the experimental glow curve data with ‘integral figure of merit’ values no worse than $\sim 5\%$. The ratio of peak 5a/5 deduced from the deconvolution procedure is, however, quite sensitive to the chosen temperature difference between peaks 5a and 5. The subtraction procedure employed to determine the peak-shape parameters of peak 5a does not determine the $T_{\max}$ of peak 5a to better than $\pm 2^\circ\text{C}$. For the results shown in Table 2, $\Delta T_{\max(5a \rightarrow 5)}$ was kept constant at 12°C . Allowing $\Delta T_{5a \rightarrow 5}$ to vary by $\pm 1\text{--}2^\circ\text{C}$ in either direction changes the values of 5a/5 significantly, but the trend and magnitude of the increase of 5a/5 with increasing ionisation density, linear energy transfer (LET) are maintained. The temperature ‘jitter’ in the value of $T_{\max}$ of composite peak 5 of 1.75°C and the uncertainty in the temperature difference between peaks 5a and 5 are thus the major factors in determining the precision of measurement of the ratio of peak 5a/5 induced by the various radiation fields.

RESULTS AND DISCUSSION

Table 2 summarises the experimentally measured peak 5a to peak 5 relative intensities in a large variety of LID and HID radiation fields including (i) $^{90}\text{Sr}/^{90}\text{Y}$ beta rays, (ii) 0.1–1.5 keV monoenergetic electrons, (iii) low-energy X rays, (iv) protons and deuterons, (v) He ions and (vi) Cu and I ions. The ratio of the intensities of peak 5a to peak 5 is increased in the slow-cooled samples when compared with the naturally cooled samples by a factor of ~ 3 with the notable exception of the results for the low-energy proton irradiations, which show an even greater increase. This increase results in significantly improved precision in the measurement of 5a/5 with an SD averaged over all the radiation fields of 15 % compared with 25 % for the naturally cooled samples. The peak 5a/5 ratio in the slow-cooled material for the various radiation fields is shown in Figure 4 plotted against LET, which serves as a rough indicator of ionisation density. The data for each radiation species clearly show an increase in

Table 1. Glow peak parameters—first-order kinetic analysis.

Glow peak	$T_{\max}$ ($^\circ\text{C}$)	Activation energy (eV)
2	119	1.10
3	162	1.15
4	182	1.90
5a	196	2.30
5	208	2.15
5b	224	2.25
6	242	2.1
7a	258	2.00
7	275	2.00
8	300–310	2.00
9	320–330	2.8
10	348	2.8

Table 2. Ratio of peak 5a to peak 5 as a function of ionisation density and cooling rate.

Radiation	Energy (MeV)	(LET) _{ave} (keV μm^{-1}) in water	Fluence/ dose (cm^{-2})/ (Gy)	Cooling rate (K h^{-1})		Increase in 5a/5 ratio ^a
				100	1000	
[e ⁻] ^g	~0.5	(~5.6)	0.5	0.13 ± 0.02	0.04 ± 0.02	3.2
[e ⁻]	1500 × 10 ⁻⁶	10		0.95 ± 0.2		
[e ⁻]	1000 × 10 ⁻⁶	13		1.02 ± 0.09		
[e ⁻]	500 × 10 ⁻⁶	19.5		1.12 ± 0.07		
[e ⁻]	100 × 10 ⁻⁶	30		1.27 ± 0.07		
[e ⁻] ^b	100–1500 × 10 ⁻⁶ (no significant variation with energy detected)				0.61 ± 0.08	
X rays ^c	0.069		1.0		0.35 ± 0.05	
X rays ^c	0.153		1.0	0.20 ± 0.015		
X rays ^c	0.040		1.0	0.19 ± 0.014	0.089 ± 0.027	2.1
X rays ^d	0.028		0.4	0.18 ± 0.020		
X rays ^d	0.097		1.0		0.10 ± 0.016	
X rays ^d	0.097		250		0.07 ± 0.03	
X rays ^e	0.1		1.0		0.12 ± 0.04	
Protons ^d	4.5	9	10 ⁹	0.27 ± 0.036	0.14 ± 0.057	1.9
Deuteron ^d	4.5	15	10 ⁹	0.31 ± 0.11	0.10 ± 0.02	3.1
Protons ^f	2.6	13	1.26 × 10 ¹⁰	0.54 ± 0.18	0.08 ± 0.09	6.8
Protons ^d	1.3	22	10 ⁹	0.43 ± 0.016	0.096 ± 0.027	4.5
Deuteron ^d	1.7	30	10 ⁹	0.58 ± 0.12	0.23 ± 0.05	2.5
Alpha ^d	7.5	67	10 ⁹	0.35 ± 0.08	0.12 ± 0.03	2.9
Alpha ^d	2.0	150	10 ⁹	0.69 ± 0.14	0.37 ± 0.14	1.9
Cu ^{+3f}	20.6	406	4 × 10 ⁸	0.78 ± 0.13	0.33 ± 0.095	2.4
I ^{+10f}	82.5	711	2.3 × 10 ⁹	0.60 ± 0.08	0.27 ± 0.08	2.2
I ^{+10f}	20.7		4 × 10 ⁹	0.80 ± 0.04	0.35 ± 0.07	2.3

^aOwing to slow cooling compared to fast cooling.

^bElectron gun: Tel Aviv University.

^cXRAD 320 X-ray tube: Westmead Millenium Institute, Sydney, Australia.

^d5 MeV Singletron (High-Voltage Engineering) Radiological Research Accelerator Facility, Columbia University, USA.

^eEuropean Synchrotron Radiation Facility, Grenoble, Switzerland: full width at half maximum of the energy spectrum=70 keV.

^f10 MeV ANTARES Tandem, Australian National Science and Technology Organization, Sydney Australia.

^g⁹⁰Sr/⁹⁰Y source: Ben Gurion University, Israel.

the peak 5a/5 ratio with decreasing particle energy (increasing ionisation density). It deserves mention that although the ratio of the intensity of peak 5a to peak 5 increases, the overall relative TL efficiency of composite peak 5 (normalised to LID radiation) decreases owing to full population of the available TCs and LCs in the high-dose regions in the track where the radial dose is greater than ~1000 Gy). Two competing effects of ionisation density are thus at play: increased geminate/localised recombination (peak 5a) owing to an increase in the relative number of e-h occupied TC/LC pairs and decreased TL efficiency owing to full occupation of the TCs and LCs leading to dose 'wastage'⁽²³⁾. The value of the 5a/5 ratio for the very heavy ions (Cu and I) is somewhat lower than expected and may indicate the role of defect production owing to enhanced nuclear scattering in the efficiency of the luminescence

mechanisms⁽³⁾. Representative deconvoluted glow curves for the slow-cooled materials are shown in Figures 5–11 indicating values of 5a/5, which increase by an order of magnitude, from 0.13 ± 0.03 following ⁹⁰Sr/⁹⁰Y irradiation to 0.43 ± 0.016 for 1.3 MeV protons to 0.80 ± 0.04 for 20.7 MeV I⁺¹⁰ ions and finally to 0.95–1.27 for low-energy electrons. Although track structure model calculations⁽¹³⁾ can describe the basic underlying mechanisms leading to increased values of 5a/5 following HCP irradiation, the many unknowns preclude, at this stage of knowledge, detailed modelling of these experimental results. Nonetheless, as the HCPs studied herein produce secondary electrons of maximum energy of a few kiloelectron volts⁽²⁴⁾, the 5a/5 values induced by low-energy electrons might be expected to reflect this fact and lead to values close to those following HCP irradiation. The glow curve shown in

Figure 4. Intensity ratios of 5a/5 as a function of ionisation density/LET for different charged particle species.

Figure 5. Deconvoluted glow curves following $^{90}\text{Sr}/^{90}\text{Y}$ irradiation: naturally cooled material (right); slow-cooled material (left).

Figure 11 for 1.5 keV electrons is rather anomalous with more intensity in the high-temperature region than expected. The electron fluence was purposely limited so that estimation of the electron fluence/dose based on the measured TL intensity, electron ranges in LiF of ~ 1000 Å and expected values of the relative TL efficiency for low-energy electrons indicate that the anomaly is not significantly affected by the dose. Similar anomalous results have been

reported by Carrillo *et al.*⁽²⁵⁾ following irradiation by low-energy photons who recommended that annealing be carried out in an inert gas to minimise surface effects. At least three other issues may be involved in the results following low-energy electron irradiation. The first is the radial dose distribution in the HCP tracks that varies by ~ 10 orders of magnitude from the track 'core' to the periphery of the track, thereby leading to radial distributions of the

Figure 6. Deconvoluted glow curve following 4.5 MeV proton irradiation in a slow-cooled sample.

Figure 7. Deconvoluted glow curve following 7.5 MeV alpha particle irradiation in a slow-cooled sample.

TC, LC and competitive centre occupation probabilities very different from those following the low dose-electron irradiation⁽¹³⁾. The second, as previously mentioned, is the possibility of enhanced defect production owing to nuclear scattering following HCP irradiation^(24, 26). And, the third is the possibility of surface effects influencing the TL mechanism following irradiation by low-energy electrons in samples annealed in air⁽²⁷⁾. Studies using ultra-soft synchrotron X rays in both nitrogen and

air-annealed samples are planned in order to resolve the latter possibility.

CONCLUSIONS

The shape of composite Peak 5 is dependent on ionisation density owing to enhanced localised recombination at HID, which results in an increase in the relative intensity of a low-temperature satellite, peak 5a. The increased relative intensity of peak 5a in the

Figure 8. Deconvoluted glow curve following 1.7 MeV deuteron irradiation in a slow-cooled sample.

Figure 9. Deconvoluted glow curve following 2.0 MeV alpha particle irradiation in a slow-cooled sample.

HID-induced glow curves broadens composite peak 5 and ‘pulls’ the value of $T_{\max}$ of composite peak 5 to slightly lower temperatures. The ratio of peak 5a to peak 5 determined by computerised glow curve analysis is found to increase by ~ 1 order of magnitude from the glow curves induced by LID radiation fields to the HID radiation fields. Significantly improved precision of measurement of the ratio of peak 5a/5 has been achieved for a large variety of radiation fields in slow-cooled samples and using peak-shape parameters of

peaks 4 and 5 based on ancillary measurements in ‘single-peak’ glow curves following LID irradiation. The systematic increase in the ratio of 5a/5 by ~ 1 order of magnitude with increasing ionisation density demonstrates the viability of the concept of the peak 5a/5 nanodosimeter. The precision of measurement depends on strictly applied protocols both in the preparation/annealing protocols of the material and in the deconvolution analysis as the ratio of peak 5a/5 is strongly dependent on the chosen temperature

Figure 10. Deconvoluted glow curve following 20.6 MeV copper ion irradiation in a slow-cooled sample.

Figure 11. Deconvoluted glow curve following 1.5 keV electron irradiation in a slow-cooled sample.

difference between peaks 5a and 5. The results shown above are, therefore, not easily transferable to other measurement modalities and should not be attempted to be reproduced unless great care is taken to reproduce all the readout and analysis and experimental conditions. These results are primarily intended to demonstrate the concept and potential of a solid-state nanodosimeter based on the existing LiF:Mg,Ti materials. Further work is necessary to improve the reproducibility of the heat transfer to the sample during readout in order to improve the precision in the appearance of $T_{\max}$ of peak 5 from $\pm 2^\circ\text{C}$

to $\sim \pm 0.5^\circ\text{C}$. Such an improvement would be expected to profoundly improve the precision of measurement of the 5a/5 ionisation density-dependent nanodosimetric signal.

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