Jurnal Sains Nuklear Malaysia, 2018, 30(2): 1 – 14
ISSN: 2232-0946

ASSESSMENT OF THERMOLUMINESCENCE GLOW CURVES AND KINETIC PARAMETERS OF FABRICATED GE-DOPED FLAT FIBER FOR RADIOTHERAPY APPLICATION

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ABSTRACT

The thermoluminescence kinetic parameters of Ge-doped flat fibre have been investigated comprehensively using the computerized glow curve deconvolution analysis. The Ge-doped flat fibre was irradiated to 6 MV and 10 MV photon beam with dose ranging from 100 cGy to 300 cGy. Analysis was done using WinGCF software on the dependence of the glow curve intensity on dose delivered and the determination of the trap parameters. Ge-doped flat fibre was found to be linear over the entire dose range explored for both 6 MV and 10 MV irradiations with r^2 value of 0.9955 and 0.9933 respectively. The glow curve consists of five individual glow peaks. The peak height increases with increasing irradiation dose. The first peak (P1) is a dominant individual peak for low temperature peak (LTP) with the maximum temperature ranging from 167.4°C to 179.0°C for both beams studied. Meanwhile, peak (P4) is a dominant individual glow curve for high temperature peak (HTP) with maximum temperature for 6 MV and 10 MV and is observed between 264.5°C to 279.4°C. Peak 1 has the lowest activation energy which is less than 0.72 eV while peak 2 shows the highest activation energy (1.3 eV < Ea < 2.1 eV) which indicates the deepest electrons trap. The results showed that the peak integral will increases as the dose increases. The Ge-doped flat fibre demonstrated the second-order kinetic peak behavior by exhibiting the symmetric shape of the glow curve with high temperature half of the curve slightly broader than the low temperature half, which suggests the possibility of electron retrapping.

Keywords: Flat fibre, Ge-doped, glow curve deconvolution, kinetic parameters

INTRODUCTION

Latest research developments in the optical fibre sensor technology have led to a renewed interest in dosimeter for radiation detection. The idea turns up when the silica-based optical fibre possesses thermoluminescence (TL) characteristics as the famous phosphor-based dosimeter. Sharing a common basic fundamental, the aim is to introduce novel fabricated optical fibres as a new potential thermoluminescent material as the medium that's been invented not only can be used as dose detector but it must also be easy to be implemented in clinical practices. A lot of research groups globally have looked into the TL response of silica dioxide (SiO_2) optical fibres for radiation dosimeter. All of those studies recognized the capability of this optical fibre in radiation detection for various types of radiation such as gamma (Entezam et al., 2016a; Nawi et al., 2015; Wahib et al., 2015), photons (Entezam et al., 2016b; Noor et al., 2014; Zahaimi et al., 2014), electrons (Alawiah et al., 2013; Hashim et al., 2015; Wagiran et al., 2012), protons (Hassan et al., 2017) and neutron (Hashim et al., 2010). Without any doubt, the ability showed by optical fibre as potential radiation sensor has infiltrated a new boundary wherein the traditional optical fibre only exploits the field of telecommunication technology.

Previously, a phosphor crystalline element such as Lithium Fluoride (LiF) dominates the use in thermoluminescence dosimetry (TLD) field before the introduction of optical fibre. However, several drawbacks exhibited by this material including potential hygroscopic problem, expensive and low spatial resolution make them a secondary choice in radiation dosimetry. These limitations of the phosphor TLD have stimulated research of numerous alternative materials for medical radiation TL dosimeters particularly using optical fibre with improved characteristics. These interests nevertheless have incited the ongoing exploration of doped optical fibres as radiation dosimeter. A lot of research consortiums have looked into the TL response of silica optical fibres doped with germanium (Ge) for application as radiation detector. Some reviews (Bradley et al., 2012; O'Keeffe et al., 2015; Yusoff et al., 2005) appeared to support the facts that the Ge-doped optical fibers provided promising response in ionizing radiation dosimetry in terms of good linearity and reproducibility, angular and dose-rate independence plus low fading effect typically in radiotherapy dose range (Alawiah et al., 2015a; Moradi et al., 2017; Noor et al., 2014). Besides those favorable properties, it also has additional advantages than conventional dosimeters, which are: good spatial resolution due to its small sizes (few microns), impervious to water, chemically inert and modest cost (Abdulla et al., 2001).

In order to increase TL performance of the optical fibre, extensive researches were conducted all around the globe including manipulating the dopant concentrations (Fadzil et al., 2017; Noor et al., 2016) and sizes of the optical fibre (Begum et al., 2015; Mahdiraji et al., 2017). Nevertheless, the explorations to improve the TL capacity for optical fibre in radiation dosimetry are rapidly flourishing. Recently, several researchers have come up with a new approach of a novel form of optical fibre in which it is fabricated as flat dimension as compared to the common cylindrical shape of the commercially available optical fibre. The theoretical aspect of flat optical fibre has denotes a new defect on their collapsed-surface which significantly increased the TL sensitivity of the flat fibre (Ghomeishi et al., 2015). The flat optical fibre made of silicon-based glass, which has an amorphous structure. The silicon bonds form a continuous random network with some silicon atoms left in amorphous material which lead to increase of defects, while in regular crystalline structure, the bonds form only four bonds with surrounding atoms (Bradley et al., 2017).

The TL properties of silica based optical fibre are based on similar trapping processes as in crystalline structure. Different trap levels in the band gap of the material represent the characteristics of the glow curve. These traps are characterized by several kinetic parameters. The behaviour of amorphous structures is less well understood as compared to crystalline materials and most studies in the field of radiation dosimetry have only focused on the dosimetric characteristics of optical fibres and its clinical approaches. Researchers have not explored TL glow curve kinetic parameters in much detail for their material characterizations. This research work is mainly to analyze glow curve formation in order to examine their effect of TL response and kinetic parameters for better understanding on TL mechanism in Ge-doped flat fibre.

MATERIALS AND METHODS

Fibre Fabrication and Sample Preparation

In this study, Ge-doped flat fibre was originated from an un-collapsed hollow preform that was fabricated using a standard modified chemical vapour deposition (MCVD) process at temperature ranging from 1600ºC to 2000ºC. The process was done by introducing the silica tetrachloride ($SiCl_4$) with germanium tetrachloride ($GeCl_4$) and oxygen (O_2) along the capillary region of silica glass tube. The preform was moved forward and backward on the burner with elevated temperature while being rotated in axial rotation. The whole procedure of perform fabrication used the same methodology as detailed by Noor et al. (2016).

The un-collapsed Ge-doped preform later was being made to a novel form of fibre in flat shape, known as flat fibre. To fabricate flat fibre, the un-collapsed Ge-doped preform was drawn down into fibre cane, of size 3 to 4 mm diameter. Upon drawing of the fibre cane, a suitable vacuum pressure (based on the size of the flat fibre) was applied from the top of the fibre cane to collapse the tube shape cane into flat shape in the hot-zone area of the furnace as shown in Figure 1. In this case, ~ 20 kPa vacuum pressure was applied to the inner part of the un-collapsed preform. At temperature of 2100°C, the fibre-drop occurred to initiate the drawing process of flat fibre.

Then, a fibre capillary cane was pulled from the bottom of the furnace at a temperature of 2000 ± 10°C with a constant drawing speed of 1 to 2 m/min. The output fibre was a flat shaped fibre having a Ge-doped core in the collapsed region. The end product of this fibre drawing was a flat fibre with dimension of 273 x 67.5 µm. The structure and the element compositions of the flat fibre were studied using a scanning electron microscope with energy dispersive x-ray fluorescence (SEM-EDX) capability.

The Ge-doped flat fibres were cut into 6.0 ± 1.0 mm lengths using a S90R diamond cutter (Thorlabs, USA) and cleaned using methanol to remove any possible deposition on the surface of the fibre. Thermal annealing was carried out in order to remove any background signal and stabilize the sensitivity. In this study, the flat fibres were annealed using a furnace (Carbolite, Derbyshire, UK). The fibres were placed on a brass plate, wrapped in aluminium foil and annealed at 400^{o}C for 1 hour. The fibres were then left inside the furnace for 10 hours to cool down to room temperature. Ten (10) pieces of the flat fibres were placed in each plastic capsule to provide a mean TL reading and variation at the chosen dose delivered.

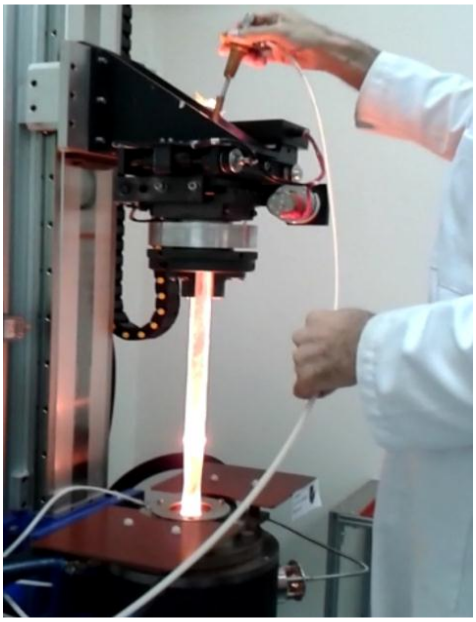

Figure 1: Vacuum pump is placed on the top of the fibre cane.

Irradiation and TL Readout

Exposure to source of radiation was done using Varian Clinac linear accelerator at Universiti Malaya Medical Centre. The plastic capsules containing flat fibres were placed on top of 10 cm solid–water™ phantom (Gammex, U.S.A) (30 cm x 30 cm) and sandwiched with 1.5 cm and 2.5 cm bolus for 6 MV and 10 MV irradiations, respectively to provide maximum dose to the samples. The flat fibres were irradiated with dose ranging from 100 cGy to 300 cGy at dose rate of 600 MU/min with source to surface distance (SSD) of 100 cm and field size of 10 x 10 cm^2.

For the purpose of TL evaluation, Harshaw 3500 (Thermo Fisher Scientific Inc, U.S.A) reader located at Secondary Standard Dosimetry Laboratory (SSDL), Malaysian Nuclear Agency was used. The nitrogen gas was introduced into the reader in order to provide oxygen-free environment, thus reducing triboluminescence effect. During TL readout, the following time temperature profiles were used: a preheat temperature of 120°C for 10 s, a readout temperature of 400°C for 13.3 s, and a heating rate of 30°C/s. Finally, an annealing temperature of 400°C was employed for 10 s to eliminate any remaining signal.

Glow Curves Analysis Using Computerized Glow Curve Deconvolution (CGCD)

The glow curves were extracted from WinREMS software as a result of heating the TL materials by the TLD reader and analyzed using WinGCF, a curve fitting software. WinGCF is a program for glow curve deconvolution with a specific purpose for TL dosimetric evaluation which fits single glow curve peak, even for low glow curves it provides approach for polishing the glow curves and diminishing background curves. Technically, in order to figure out the glow peak kinetic parameters such as the maximum peak temperature (T_{max}), peak integral (*PI*), activation energy (E_a) and frequency factor, the experimental glow curves were deconvoluted into 5 individual peaks using this software.

RESULTS AND DISCUSSION

Flat Fibre Structure

The scanning electron microscope (SEM) was carried out over the cross-sectional area of the flat fibre and was used for elemental analysis if an X-ray spectrometer is available. Interestingly, the scanning image of flat fibre (Figure 2) demonstrated a well define flat dimension of optical fibre with a bulging region on both ends of the flat fibre, similar pattern with the thinly core region. It seems possible that these results are due to the edges of the flat fibre have reached their maximum tension ability to collapse inward.

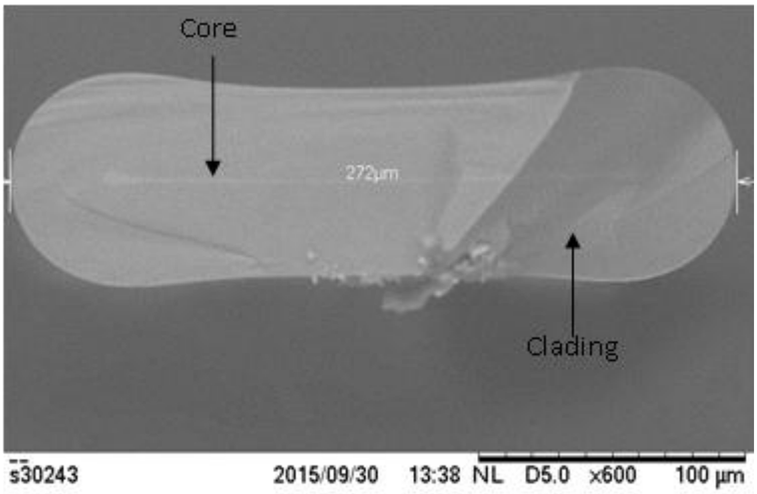

Figure 2: A cross sectional image of Ge-doped flat fibre obtained using a scanning electron microscope with energy dispersive x-ray fluorescence (SEM-EDX).

A line scanning was done at a central point of the flat fibre (Figure 3a) in order to acquire the germanium concentration profiles. Figure 3(b) illustrated that the highest germanium concentration at the core region of the flat fibre in correlation with the diffusion of the germanium during the MCVD process.

TL Performance

From the data obtained, it is clearly observed that Ge-doped flat fibre showed a linear response for the dose range of 100 cGy to 300 cGy with r^2 value of 0.9955 and 0.9933, for 6 MV and 10 MV photon irradiations, respectively (Figure 4). It is also found that the TL signal of the flat fibre irradiated by 10 MV is 11.7% higher, as compared to with 6 MV photon beam. These results are in accordance with recent studies by Bradley et al. (2015) and Ghomeishi et al. (2015) who reported that TL intensity of flat fibre will increases as the energy of the irradiation increases.

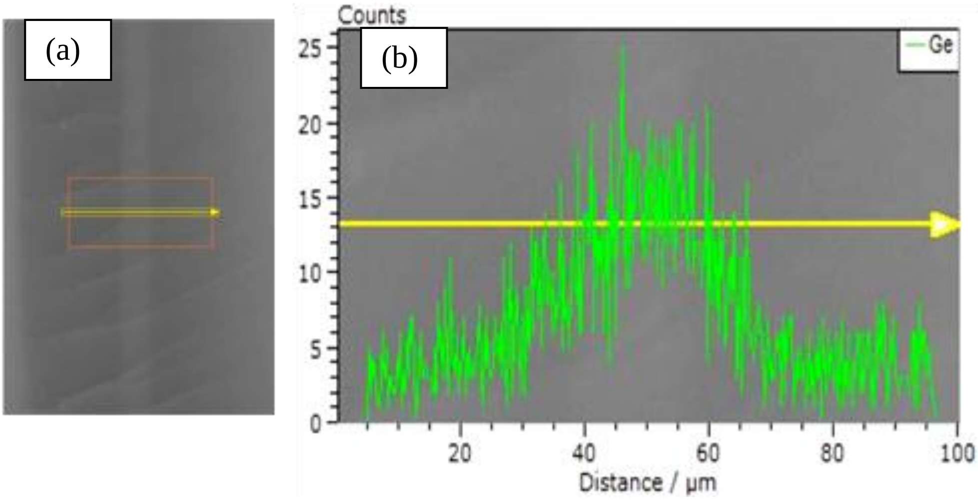

Figure 3: Line scan analysis (a) that included the core and cladding of the flat fibre and the distribution of germanium (b) along the line scan area showed the accumulation of dopant element at the central region of flat fibre.

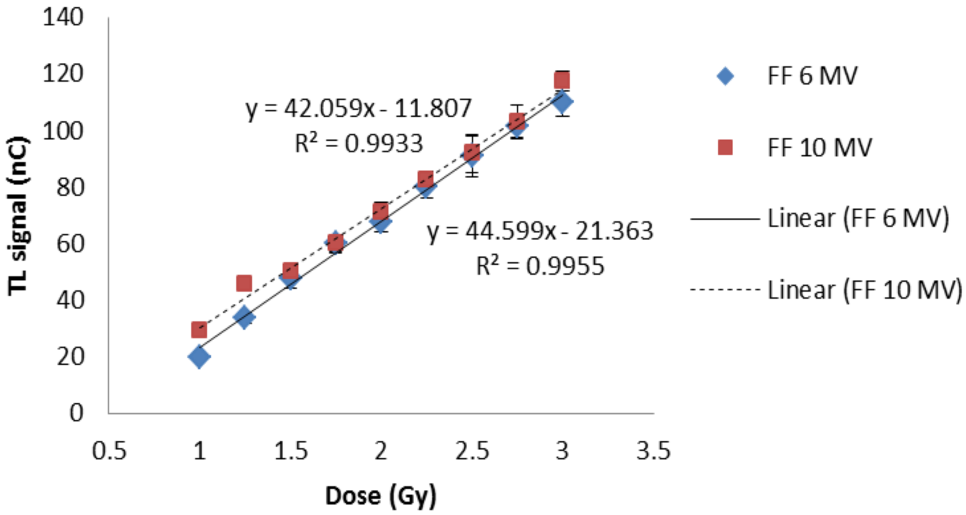

Figure 4: Linearity dose response of Ge-doped flat fibre subjected to 6 MV and 10 MV photon beams.

Glow Curves and Kinetic Parameters Analysis

The most interesting finding was that the glow curves of Ge-doped flat fibres appeared to be a broad double peaks when exposed to the studied radiotherapy dose range which in agreement with Ghomeishi et al. (2015). The first glow peak (Channel 1-100) is denoted as a low temperature peak (LTP) while the second peak is denoted as a high temperature peak (HTP) as shown in Channel 101-200 (Figure 5(a) and (b)). It is clearly seen the peak height and the maximum temperature of the glow curve are highly dependent on the dose (Moscovitch and Horowitz, 2007). The maximum temperature of the glow curve refers to the point of the highest peak height.

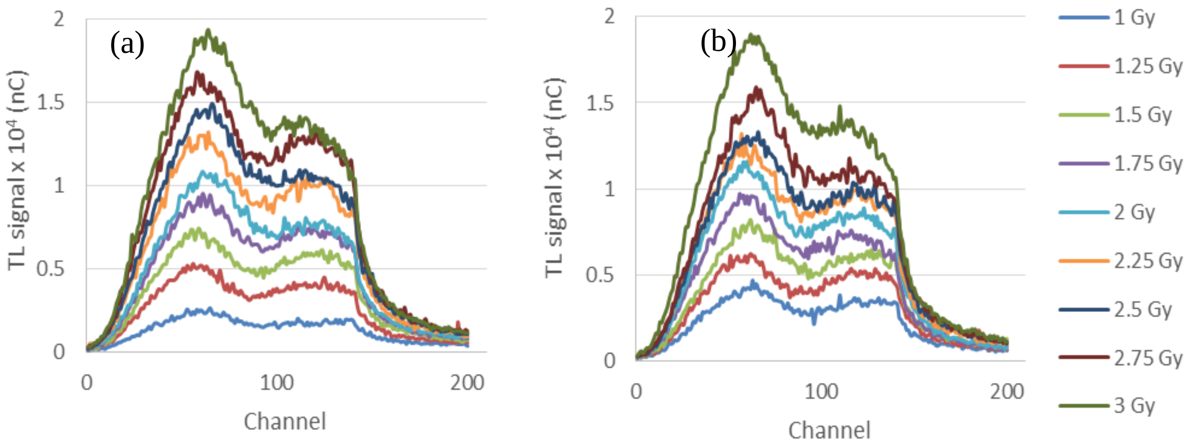

Figure 5: Typical glow curve formation of Ge-doped flat fibre subjected to 6 MV (a) and 10 MV (b) photon irradiations.

The LTP is observed with maximum intensity at 231°C to 251°C for both 6 MV and 10 MV irradiations. Meanwhile for the HTP, the maximum peak height is observed at 327°C to 368°C and 338°C to 372°C for 6 MV and 10 MV photon beams, respectively. However, the findings of the current study do not support the previous research on Ge-doped flat fibre by Alawiah et al. (2015a) where the glow curve of Ge-doped flat fibre comes out as a single peak curve. A possible explanation for this might be that the previous study makes used of ultra-high dose from 1 kGy up to 1000 kGy in which the electrons occupied more traps in higher dose compared to the lower dose.

It is also found that the shape of the glow curve is constant with increasing dose. As the dose increases, area under the glow curve increases, suggesting an increasing number of electrons released from its traps. These findings suggest that in general the Ge-doped flat fibre demonstrates the second-order kinetic peak as proposed by Garlick and Gibson (1948) which stated that the shape of the glow curve is nearly symmetric with the high temperature half of the curve, and it is slightly broader than the low temperature half. These results indicate the possibility of strong electron retrapping and thus causing a delay in luminescence emission. According to this model, the TL intensity, *I*, at a constant temperature can be written as in Equation 1:

$$I(T) = s\, n_o\, e^{-\frac{E}{kT}} \left[1 + \frac{s}{\beta}\int_{T_o}^{T} e^{-\frac{E}{kT'}}\, dT'\right]^{-2} \qquad (1)$$

where, s represents the frequency factor of trapped electrons, n_o is the trap's initial concentration, E is an activation energy in eV, T is temperature in unit of Kelvin (K) while β is the heating rate and k is the Boltzmann constant.

The computerized glow curve deconvolution (CGCD) analysis was used in determining a complicated thermoluminescence glow curve into individual peak elemental. Figure of merit (FOM) for deconvoluted main glow curves for each of the fiber samples was within 3.0% to 5.0%. The results revealed that the Ge-doped flat fibre glow curve consists of 5 individual peaks. It is found that the first peak (P1) is a dominant individual peak for low temperature region as shown in Figure 6(a) and (b), with the maximum temperature (T_{max}) ranging from 170.1°C to 176.8°C and 167.4°C to 179.0°C for 6 MV and 10 MV, respectively. Variation of T_{max} for glow curve peak of Ge-doped flat fibre was summarized in Table 1. In this study, P1 is classified as low temperature peak (LTP). Meanwhile, peak 4 is found to be dominant at high temperature region and has been denoted as the high temperature peak (HTP). The T_{max} for 6 MV beam was observed at 264.5°C to 272.8°C while 267.6°C to 279.4°C for 10 MV photon irradiation. These results showed that peak 1, 2, 3, 4 and 5 to overlap each other at channel 100, which is the central region of the glow curve.

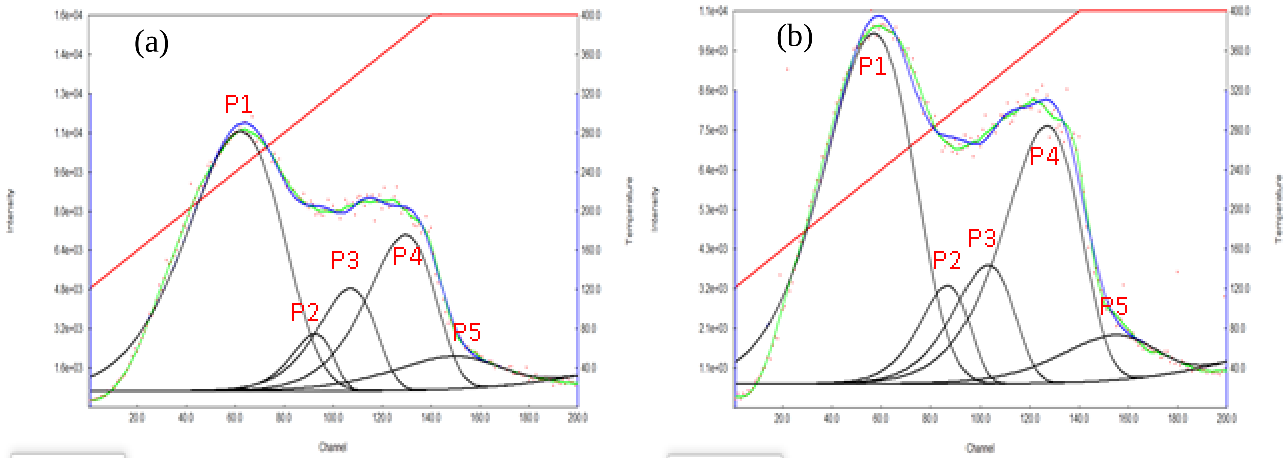

Figure 6: Glow curve deconvolution of Ge-doped flat fibre subjected to 200 cGy 6 MV (a) and 10 MV (b) irradiations

The T_{max} of the glow peaks (P1 to P5) pattern was found to be consistent throughout the dose range used for 6 MV and 10 MV in this study as shown in Figures 7(a) and (b) respectively. It is clearly indicated that the T_{max} is dose independent. Every individual deconvoluted peak (P1 to P5) required a constant temperature in order to provide the TL intensity.

Figures 8(a) and (b) showed the activation energy (E_a) of individual deconvoluted peak for both 6 MV and 10 MV photon beams, which was affected by increasing dose. The activation energy refers to the amount of energy needed to excite the electron from valence band to conduction band (Horowitz and Yossian, 1995). Peak 1 has the lowest activation energy which is less than 0.72 eV for both 6 MV and 10 MV while peak 2 required highest activation energy (1.3 eV < E_a < 2.1 eV). There is a decline trend for activation energy in peaks 3, 4 and 5 in every dose given regardless of the photon energy irradiation. These results indicate that the electrons in peak 1 occupied the shallowest trap while electrons were trapped in the deepest trap in peak 2.

Table 1: Variation of maximum temperature (T_{max}) for glow curve peaks of Ge-doped flat fibre

Peak Number	**T_{max} (°C)**	
	6 MV	**10 MV**
P1	170.07 to 176.79	167.40 to 179.00
P2	209.72 to 219.71	203.52 to 216.59
P3	230.75 to 241.80	231.41 to 243.57
P4	264.50 to 272.78	267.56 to 279.43
P5	286.95 to 309.41	289.62 to 308.43

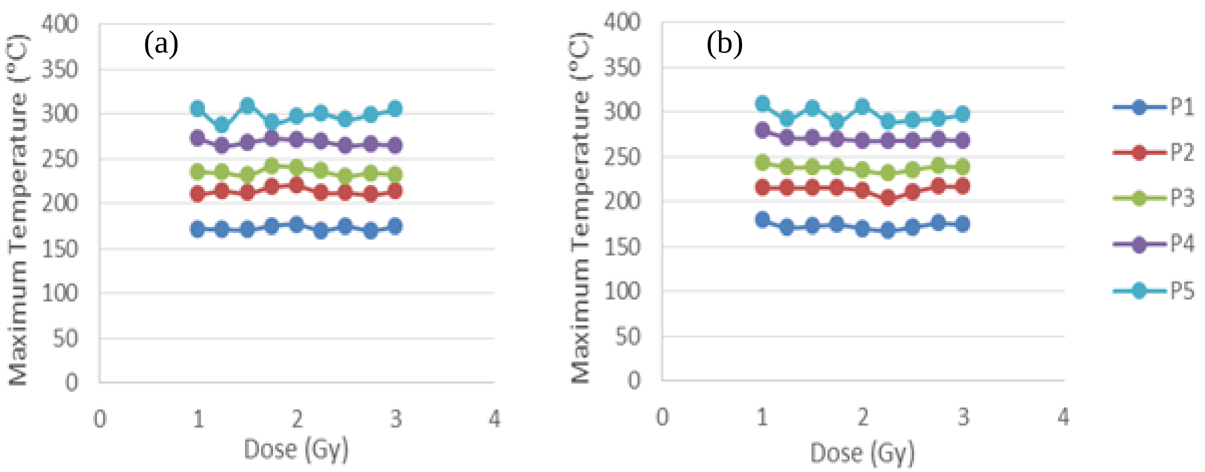

Figure 7: The influence of various doses of 6 MV (a) and 10 MV (b) irradiations on the maximum temperature

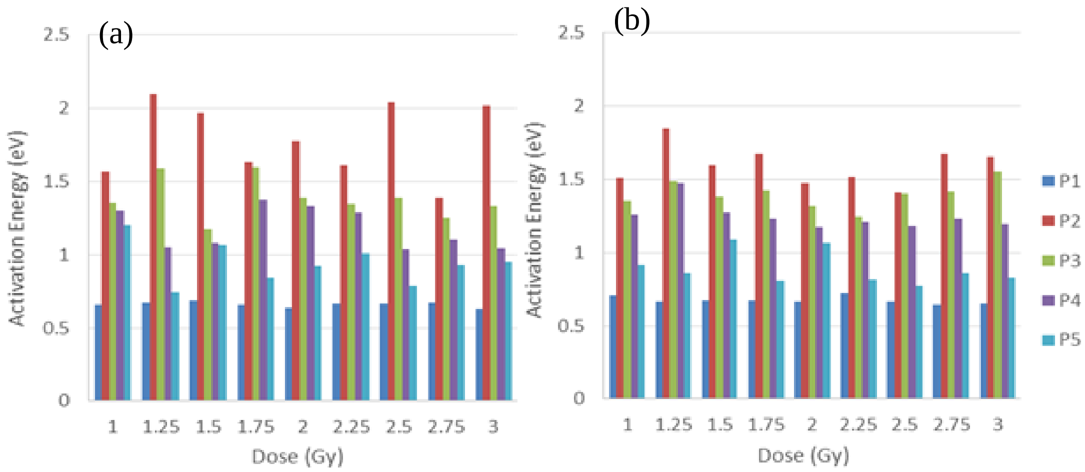

Figure 8: The influence of various doses of 6 MV (a) and
10 MV (b) irradiations on the activation energy

Figures 9(a) and (b) showed the influence of various dose on the peak integral of individual deconvoluted glow curve. The peak integral (*PI*) is also related to the peak area where it represents the amount of free electrons that travel from the traps to the recombination center (Alawiah et al. 2015b). Peak 1 has the highest *PI* value for 6 MV and 10 MV which are around 6.428 nC to 53.66 nC and 9.683 nC to 52.13 nC, respectively. The lowest *PI* value demonstrated by peak 5 indicates the smallest area under the individual curve occupied by the electrons. The results of this study showed that the *PI* was affected by the dose delivered. As the dose increases, the *PI* increases. In general, *PI* value for 6 MV is 14% higher as compared to 10 MV photon beams.

Figures 10(a) and (b) showed the relationship between T_{max} and E_a. The results demonstrated in general that peak 2 (1.3 eV to 2.1 eV), peak 3 (1.1 eV to 1.6 eV), peak 4 (1.0 eV to 1.5 ev) and peak 5 (0.7 eV to 1.2 eV) obey the second order kinetic where the E_a is constantly increased with increasing T_{max} (Garlick and Gibson, 1948). In contrast, the temperature has minimal effect on the value of E_a for peak 1 (0.6 eV to 0.7 eV). The result is applicable for both 6 MV and 10 MV photon beams. The response for Ge-doped flat fibre in this study is contradicted with Erbium-doped fibre data particularly for peak 2 and 3 as mentioned by Alawiah et al. (2015c).

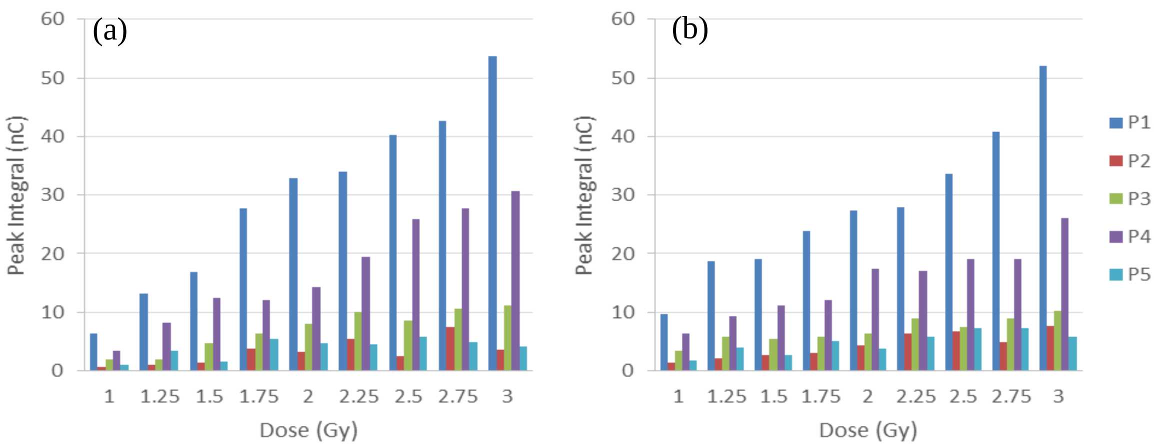

Figure 9: The influence of various doses of 6 MV (a) and
10 MV (b) irradiations on the peak integral

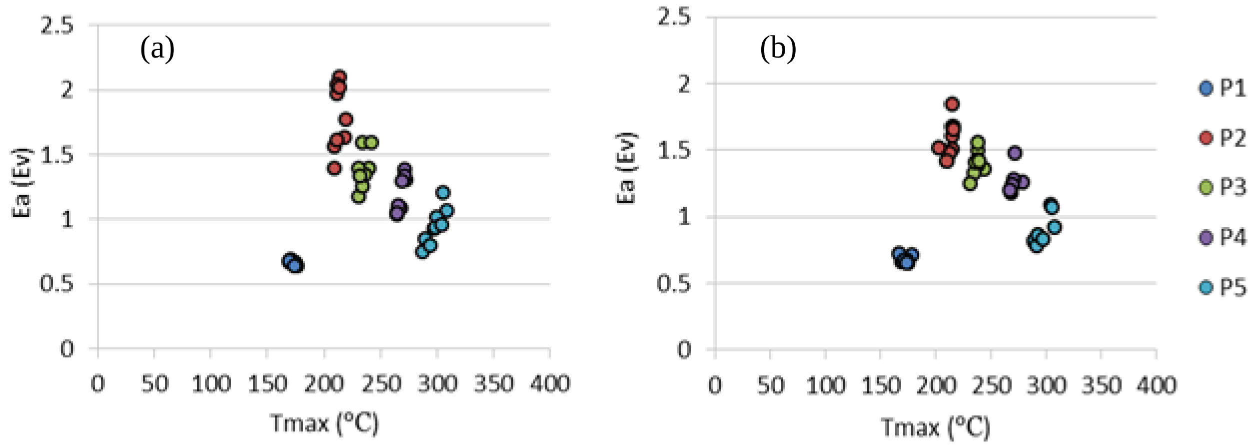

Figure 10: The variation of kinetic parameter of E_a as the function of T_{max} exposed to 6 MV (a) and 10 MV (b) photon irradiations

CONCLUSIONS

TL peaks intensity dispenses information regarding the electron-trap distribution in the TL material. In general, the glow curve of Ge-doped flat fibre consists of five individual glow peaks with the peak height increase with increasing irradiation dose. P1 is dominant individual peak for low temperature peak (LTP) while P4 has greatest influence in high temperature peak (HTP). It clearly indicates that the kinetic parameters analysis of Ge-doped flat fibre obeys the second order kinetic model where it considers the case of strong electron retrapping. High probability of retrapping is suggested due to additional defects generated by collapsing inner wall of the flat fibre during the drawing process.

ACKNOWLEDGEMENTS

This research is supported by Universiti Putra Malaysia, Geran Putra Incentive Putra Siswazah (UPM-GP-IPS) no: 9433970, Malaysia Fundamental Research Grant Scheme (FRGS) no: 5524789 and University of Malaya High Impact Research Grants (UM-HIR) no: UM.C/625/1/HIR/33. The authors would like to express their gratitude to staffs at Malaysian Nuclear Agency for the use of TL reading facilities.

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