THE EFFECT OF RAPID COOLING ON THE RELATIVE SENSITIVITY OF THERMOLUMINESCENT DOSIMETERS (TLD-100H)

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ABSTRACT:

The sensitivity of TLD is an important issue due the need of accurate dosimetry and the reuse of thermoluminescent dosimeters. Annealing procedures for many times and environment affect its sensitivity. The first step before using dosimeter is to determine relative sensitivity for dosimeters to be used, and then low sensitive dosimeters cannot be used in measuring exposure signals, so it must be excluded. This study searches how to enhance relative sensitivity for excluded TLDs to be reused and how to improve the sensitivity of the used dosimeters so its readings can be trusted. From 43 dosimeters, 4 of them had a relative sensitivity out of range, but with rapid cooling; there relative sensitivity improved to be within the range. Other dosimeters showed better sensitivity with rapid cooling, so the use of rapid cooling can improve TLD sensitivity.

KEY WORDS: thermoluminescent dosimeter (TLD), cooling, annealing, relative sensitivity.

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Introduction

The need for a wide range dosimeter with high sensitivity and excellent reproducibility pushed scientists from 1950s to do their search for a perfect TLD. Farrington Daniels and his cooperators were the first group who suggested the use of TL as a radiation dosimeter, after that, LiF:Mg was studied in 1960s and became the most important TLD material which used in many different applications such as medical dosimetry, spacecraft, mineral prospecting, environmental dosimetry, monitoring of radiation exposure, and in geological dating (Chen & Pagonis, 2011). The first TL material used as a dosimeter is TLD-100 (Shindle, Dhoble, Swart, & Park, 2012); it has a large variety of applications in radiotherapy especially in vivo dosimetry and phantom measurements because of it is free standing and the LiF:Mg.Ti has a wide dose range so it is used widely in radiotherapy (Scholz, 2006).

Advantages and Disadvantages of TLD

TLD has many advantages; it has small size, used in medical applications due to its high sensitivity for all dose values, no need for cable connections, the independence of environment conditions like temperature, and suitable for in vivo dosimetry, but it should be handled carefully because of its small size and its dose reading takes time and cannot be kept as a permanent record of dose (Kron, 1999). It can be found in many forms, has large dose range, independent on the dose rate, can be reused after annealing, low cost, quick read-out, many of them can be exposed together and by the use of calibration, 1-2% reproducibility can be achieved, but on the other side, it has many disadvantages such as losing its reading easily, only one time signal readout because it is erased during readout, it has different sensitivities, annealing cycle is needed, sensitive to light, and large dose affect its sensitivity (Savva, 2010). TLD dose range limit of detection is between 0.001 rad up to 1000 rad with high sensitivity for most radiation types (uz Zaman, Fatima, Naqvi, Parveen, & Sajjad, 2011).

Principle of TLDS

Thermoluminescent means the emission of light after heating. When ionizing radiation hits TLD material; electron will escape from one atom to another part of material and leaves a hole of positive charge behind it, then after heating the TLD, the recombination of electron and hole will occur to release energy as a visible light with intensity which is related to the amount of energy

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absorbed through exposure to radiation (HKUST, 2007). It contains a sensitive volume of 1 to 100 mg of crystalline dielectric mass with suitable activators which provide two types of centers (traps and luminescence centers). The TL process happens by ionizing radiation which elevates an electron into a conduction band to electron trap and leaves a hole that is called hole trap. These traps must be deep to prevent recombination in room temperatures (Attix, 2004). Photons produce secondary electrons by interacting with TLD and deposit their energy to produce the TLD signal (Olko, 2006).

Glow Curves

The TLD-100 contains magnesium and titanium; the magnesium increases the number of traps which increases the number of glow peaks exponentially to form the glow curves, while titanium increases the number of luminescence centers within TLD-100. It has five main peaks at room temperature, only peaks 4 and 5 are more appropriate for dosimetric purposes due to their long half life comparing with the rest (da Rosa & Caldas, 1998). The structure of glow-curve for TLD 100H has main dosimetric peak 4 which occurs at a temperature of about 220°C by activation energy (E) more than 2 eV and frequency factor higher than 10²⁰ s⁻¹ (Bilski, 2002). The main peak at its glow curve is peak 4 at about 210 °C which called the dosimetric peak that is used in dosimetric applications, other peaks 1,2, and 3 are in the range between 70 and 160°C, but peak 5 appears at ~300°C, so peaks 4 and 5 are overlapping peak. Its absorption in the range of 300 to 400 nm is defects for TL emission at peak 4 at about 200°C (McKeever, Macintyre, Taylor, McKeever, Horowitz, & Horowitz, 1993). The energy stored in the TLD crystal lattice due to the absorbed energy after irradiation is recovered by putting it on the planchet heater as a visible light. The relation between TL response and readout temperature refers to the glow curve which depends on the TL nonlinearity, dose response and annealing procedures that the longer half life peak is most stable and suitable for dosimetry (Yu & Luxton, 1999).

TLD Sensitivity

The sensitivity of TLD is the response per unit dose and varies with photon energy and with depth (Nath, Meigooni, & Meli, 1990). It is the integrated signal from the photomultiplier tube per unit of radiation exposure. The sensitivity of TL depends on the characteristic of TLD and the reader system. TLD sensitivity is independent of the various time delays between irradiation,

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prereadout anneal, and readout but was affected by the duration of annealing (Yu & Luxton, 1999). Many factors affect TLD readings, such as the stability of produced signal, the availability, fading rate which should be lower than 5% per month, glow curves must be simple, and a plain anneal heating cycle. Some activators (impurities) can be added to increase its sensitivity and efficiency by increasing the number of traps which emit more light during TL process. Ideal dosimeter has a linear response for doses which is not affected from any dose rate (IAEA, 2005).TLD-100 has sensitivity per unit mass equals 1 and its dose threshold is 50 μ Gy with fading factor of 5 per year and the energy response ratio equals 1.3 (Hufton, 1984). The relative sensitivity (RS) can be calculated for each TLD by the use of equation 1.

$$RS = \frac{TL_i}{TL_r}$$

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where $\overline{TL_r}$ is the average of TLD readings, and TL_i is TLD signal after exposing to dose D_i , while the sensitivity factor (SF) is the reciprocal of relative sensitivity and can be calculated using equation 2

$$SF = \frac{\overline{TL_r}}{TL_i}$$
 2

The response of TLD depends on the type of detector, dose level, radiation LET, annealing, activator content, and other factors (Olko, 2006).

TLD-100H (LiF:Mg,Cu,P)

The use of TLD-100H in a wide dosimetry program has a special interest because of its extreme sensitivity to maximum readout temperature and the consideration of implementation in routine dosimetry (Moscovitch, et al., 2006). The TL material has a tissue equivalent property and the (LiF:Mg,Cu,P) material has a high sensitivity and near flat photon energy response without the need to use correction factor (Lou & Rotunda, 2006). TLD-100H is an advanced TLD due to its new dosimetric material and has insignificant fade along simple glow curve structure up to one year, so it is a premier choice for dosimetry (Ramlo, Moscovitch, & Rotunda, 2007). The main difference due to components between TLD-100H and TLD-100 is that TLD-100H has ten times magnesium than TLD-100 with different concentration of other components like titanium, copper, and oxygen impurities (Bilski, 2002).

Methodology

TLD 100H is chosen in the current study because of its high sensitivity and linearity to doses, low rate of fading, with good stability through a number of readout cycles, and tissue equivalent effective atomic number (Z) to represent the contribution of photons energy. Its sensitivity is 30-40 times higher than other TLDs. The readout protocol has a maximum temperature of 300°C, but some additional traps do not appear at this temperature. During radiation; the radiation induced population of the traps above Fermi level which refers to radiation defect causes electronic excitation and displacement damage. TLD will keep this energy state after irradiation, and after a period of time it can be readout. The reuse of TLD is after annealing by a furnace to allow the mobilization of holes and electrons in traps to the equilibrium positions, and then TLD will give its glow curves by the use of Harshaw 3500® (Herr, 2010).

Annealing Procedure for TLD-100H and Sorting

The annealing of TLD-100H before irradiation is a compulsory step and consists of two main steps; the first is prior irradiation which consists of high temperature annealing and the second is after irradiation and prereadout annealing at low temperature. The procedure of annealing is done by the use of TLD annealing furnace which has maximum heating rate of 15°C/min. The followed annealing procedure for TLD 100H is 240 °C for 10 minutes before irradiation and the prereadout annealing is 100 °C for 10 minutes after 24 hours and before reading the TLD signals by the use of Harshaw[®] 3500 TLD reader (IAEA, 2013). Harshaw[®] 3500 TLD reader is used to read signals by WinREMS software from each TLD 100H with presence of nitrogen gas after 30 minutes of turning it on. The time between irradiation and readout must be the same to keep same fading from one calibration to another for all dosimeters (Abou-Elenein, Attalla, Elmoniem, Eldesoky, & Farouk, 2014). TLD 100 (LiF:Mg,Ti) must be annealed at 400 °C for 24 hours before reading signals by TLD reader (Harshaw) which can be converted to dose after calibration process (Olko, 2006).

The sensitivity test for 43 dosimeters (TLD-100H) presented by putting all TLDs inside plastic holder and exposing it five times to x-ray dose of different KVp at 100 SSD and 10×10 cm² field size as figure 2. The used TLD reading (*TL_i*) is a net reading without the effect of background

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signal. The annealing procedure followed in this study is by the use of oven to heat all TLDs to 240°C for 10 minutes immediately before irradiation. After a period of time (24 hours), all TLDs annealed to 100°C for 10 minutes and signals read by Harshaw 3500[®] TLD reader. The plastic holder was put on solid water phantom to absorb scattered radiations and give real doses. Cooling cycles were as the following description in table 1.

| Scan | Description of cooling cycle | | | | | | | | | |
|------|---|--|--|--|--|--|--|--|--|--|
| 1 | TLD tray was put at room temperature directly after annealing to 240°C | | | | | | | | | |
| 2 | TLD tray was put at room temperature directly after annealing to 240°C and with the use | | | | | | | | | |
| | of electric fan. | | | | | | | | | |
| 3 | TLD tray was put at room temperature directly after annealing to 240°C and with the use | | | | | | | | | |
| | of electric fan closer to the tray. | | | | | | | | | |
| 4 | TLD tray was put on ceramic plate on ice at 0°C temperature directly after annealing to | | | | | | | | | |
| | 240°C without fan. | | | | | | | | | |
| 5 | TLD tray was put directly on ice at 0°C temperature directly after annealing to 240°C | | | | | | | | | |
| | without fan. | | | | | | | | | |
| 6 | TLD tray was put directly on ice at 0°C temperature directly after annealing to 240°C | | | | | | | | | |
| | without fan. | | | | | | | | | |

Table 1: Description of cooling cycles for all scanned TLDs

Results

The six scanning cycles for dosimeters gave different relative sensitivity for each which calculated by the use of equation 1, and tabulated in table 2. The result was 4 of scanned TLDs had relative sensitivities out of the range (1 ± 0.15) at the first x-ray scanning. The relative sensitivities for 43 dosimeters describe their range from unity are tabulated in table 3.

| Table | 2: | Relative | sensitivity | (RS) | for | 43 | dosimeters | (TLD-100H) | underwent | 6 | cooling |
|---------|----|----------|-------------|------|-----|----|------------|------------|-----------|---|---------|
| cycles. | | | | | | | | | | | |

| TLD | RS_1 | RS_2 | RS ₃ | RS_4 | RS ₅ | RS ₆ |
|-----|----------|----------|-----------------|----------|-----------------|-----------------|
| "1 | 1.321999 | 1.182102 | 1.038862 | 1.029572 | 1.021374 | 1.041993 |

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|--------------|--------------------|-------------------------|-------------------------|------------------------|-----------|----------|------------------------|
| | "2 | 1.111466 | 1.050758 | 1.006242 | 1.046895 | 1.091468 | 1.08007 |
| | "3 | 0.953373 | 1.101275 | 0.989761 | 1.036972 | 1.046274 | 1.045941 |
| | "4 | 0.941492 | 1.081068 | 0.995512 | 0.999797 | 1.033498 | 0.971056 |
| | "5 | 1.066189 | 0.899206 | 0.881737 | 0.927879 | 1.063743 | 0.980745 |
| | "6 | 0.969747 | 1.070965 | 1.000906 | 0.994049 | 0.987818 | 1.017036 |
| | "7 | 0.980393 | 0.980034 | 1.079225 | 1.027159 | 1.0718 | 1.077028 |
| | "8 | 0.85353 | 1.020447 | 1.030303 | 1.005953 | 0.994553 | 1.008106 |
| | "9 | 1.085235 | 1.081068 | 1.069781 | 0.985546 | 1.014447 | 1.005474 |
| | "10 | 0.983227 | 0.949723 | 0.927537 | 0.921473 | 0.929987 | 0.929998 |
| | "11 | 0.965496 | 0.980034 | 0.989726 | 1.032053 | 1.005269 | 1.004641 |
| | " <mark>12</mark> | 0.890893 | 1.050758 | 1.004406 | 1.079858 | 1.010415 | 1.07 <mark>5894</mark> |
| | " <mark>13</mark> | <mark>0.8</mark> 45596 | 1.060861 | 1.051064 | 1.153409 | 1.047456 | 1.04067 |
| | "14 | 1.061392 | 1.000241 | 0.993633 | 1.000541 | 0.980117 | 1.035587 |
| | "15 | 0.892391 | 0.980034 | 0.95412 | 0.961092 | 0.896065 | 0.939613 |
| | " <mark>16</mark> | 0.975353 | 1.020447 | 1.021873 | 1.035527 | 1.074051 | 1.071731 |
| | " <mark>17</mark> | 0.998305 | 1.060861 | 1.044841 | 1.026074 | 1.002801 | 1.009025 |
| | " <mark>18</mark> | 0.924228 | 0.990137 | 1.086705 | 1.060061 | 1.042893 | 0.995892 |
| | " <mark>19</mark> | 0.912347 | 0.959827 | 1.025988 | 1.019143 | 0.958442 | 0.977723 |
| | " <mark>20</mark> | 0.940986 | <mark>0.92951</mark> 6 | 0.9 <mark>75</mark> 63 | 0.921144 | 0.933185 | 0.937162 |
| | " <mark>2</mark> 1 | 0. <mark>996</mark> 888 | 1.0406 <mark>5</mark> 4 | 1.014079 | 1.032248 | 1.039999 | 0.993119 |
| | " <mark>22</mark> | 1. <mark>032</mark> 814 | 0.889103 | 0.970165 | 0.962421 | 1.039043 | 0.998121 |
| | " <mark>23</mark> | 1. <mark>05</mark> 0746 | 0.980034 | 1.000113 | 1.040263 | 0.89889 | 0.920557 |
| | " <mark>24</mark> | 1. <mark>05</mark> 7102 | 1.050758 | 0.977609 | 1.010183 | 1.034446 | 0.985498 |
| | " <mark>25</mark> | 1.144679 | 0.919413 | 0.893339 | 0.893014 | 0.898221 | 0.894855 |
| | "26 | 1.05949 | 0.990137 | 0.975509 | 1.01417 | 0.964082 | 0.979227 |
| | "27 | 1.092501 | 0.990137 | 0.968851 | 1.008141 | 1.018332 | 1.000606 |
| | "28 | 1.022815 | 0.990137 | 0.991183 | 1.01384 | 1.046318 | 1.010899 |
| | "29 | 1.084729 | 1.010344 | 0.946033 | 0.951157 | 0.950046 | 0.99647 |
| | "30 | 0.941735 | 0.798172 | 0.828215 | 0.866884 | 0.883811 | 0.856952 |
| | "31 | 1.002009 | 1.050758 | 0.994048 | 1.016644 | 1.040842 | 1.051312 |
| | "32 | 1.148747 | 1.141689 | 1.091963 | 1.11234 | 1.089069 | 1.082709 |

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|--------------------|------------------------|----------|----------|-----------|----------|------------------------|
| "33 | 1.04769 | 0.889103 | 0.952477 | 1.008263 | 0.933272 | 0.959695 |
| "34 | 0.989764 | 1.020447 | 1.027939 | 0.990349 | 1.007181 | 0.978764 |
| "35 | 0.882008 | 0.848689 | 0.904327 | 0.830313 | 0.940103 | 0.935309 |
| "36 | 0.867961 | 1.040654 | 1.032418 | 1.034077 | 1.002184 | 1.014182 |
| "37 | 0.800684 | 1.020447 | 1.069131 | 1.045304 | 0.981803 | 0.997013 |
| "38 | 0.880955 | 0.959827 | 1.035961 | 1.014231 | 1.020627 | 1.00399 |
| "39 | 1.094748 | 0.990137 | 1.06438 | 0.979335 | 1.024494 | 1.027725 |
| "40 | 1.236405 | 0.889103 | 0.934324 | 0.861563 | 0.943519 | 0.922075 |
| "41 | 1.072666 | 1.060861 | 1.039098 | 1.031157 | 1.071087 | 1.048613 |
| "42 | 0.924896 | 0.980034 | 1.030103 | 0.972435 | 1.003288 | 1.06828 |
| " <mark>4</mark> 3 | <mark>0.89</mark> 4334 | 0.989167 | 1.090884 | 1.047468 | 0.963682 | 1.02 <mark>8645</mark> |
| | | | | | | |

| Table 3: | Relative | sensitivities | for 43 | 3 dosimeters | describe | their rang | e from | unity |
|----------|----------|---------------|--------|--------------|----------|------------|--------|-------|
| | | | | | | | 50 0 | |

| Relative sensitivity | 1 | 2 | 3 | 4 | 5 | 6 |
|--------------------------------|----------|----------|------------------------|----------|----------|------------------------|
| 1±0.05 | 14 | 21 | 28 | 32 | 28 | 28 |
| (1 ± 0.05) to (1 ± 0.10) | 16 | 14 | 12 | 6 | 11 | 13 |
| (1 ± 0.10) to (1 ± 0.15) | 9 | 5 | 2 | 3 | 4 | 2 |
| 1± (>0.15) | 4 | 3 | 1 | 2 | 0 | 0 |
| Max difference from unity | 0.321999 | 0.201828 | 0.17178 <mark>5</mark> | 0.169687 | 0.116189 | 0.14 <mark>3048</mark> |

Conclusion

The first normal relative sensitivity test for 43 TLDs appeared that 4 TLDs had low relative sensitivities out of the range (1±0.15) but by the use for rapid cooling, its sensitivity was within the accepted range. The sensitivity of 84% of TLDs was enhanced and developed by fast cooling but 16% appeared no response for this operation. The effect of rapid cooling had more effect on low sensitive TLDs more than high sensitive dosimeters, and the number of high sensitive TLDs (1±0.05) was increased from 14 up to 32. It can be said that the rapid cooling increases the sensitivity for thermoluminescent dosimeters.

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