Thermoluminescent Response of Beta-irradiated Silica Beads

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Abstract

Research into the potential for silica based materials as TLDs continues to increase, harnessing the various advantages that the materials have to offer as a dosimeter, including; affordability, chemical inertness, geometry and size, while producing suitable TL yields. Studies into the possibility of beta source dosimetry open another range of applications for this type of dosimeter, including procedures involving the possibility of *in vivo* dosimetry (Morino et al., 2002). This work aims to further investigate the properties of a much-studied batch of Mill Hill silica beads of various colours, irradiated with a $\text{Sr}^{90}/\text{Y}^{90}$ source yielding a dose rate of 0.02 Gy/s, activity 37 MBq and maximum energy of 2.27 MeV, facilitated by the Risø TL/OSL reader. The TL yield is compared to that in previous work, with the same set of beads irradiated by a 6 MV photon beam provided by a Varian electron linac (Varian Medical Systems, Palo Alto, CA) X-ray source, delivering a dose of 10 Gy. Present study shows a decrease in the mass normalised TL yield from X-ray to beta source by a factor of 10, assumed to be accounted for by the estimated mean penetration depth of the beta particles in the doped silica beads. Investigating the potential for doped silica beads to be used as a beta source dosimeter a linearity study is reported with a dose range of 5 to 20 Gy. The studies show that all coloured beads except green show a strong conformity to a linear fit of the data over the tested range of doses. Fading studies conducted with clear petite beads (producing the greatest TL yield/mg) for doses of 5 Gy and 20 Gy have shown that the majority of fading occurs within the first hour following irradiation, with a plateau in the fading to approximately 17 % and 22 % loss of signal for 5 Gy and 20 Gy respectively in the interval of 24 hours to 70 hours after irradiation. The decay of the TL yield/mg was fitted to a logarithmic trend of reduction for both the 5 Gy and 20 Gy irradiation, with R^2 values of 0.9883 and 0.9705 respectively.

Keywords: Thermoluminescence (TL), Silica, Beta, Dosimetry, *in vivo*, Risø TL/OSL

1. Introduction

Previous studies (Jafari et al., 2014a; Abdul Sani, 2014; Bradley et al., 2014) underline growing interest for the use of silica doped materials as dosimeters, most generally for medical application, with discussion of applications for *in vivo*

dosimetry (O’Keeffe et al., 2015; Mizanur Rahman et al., 2015). The applications of using silica beads as a viable dosimetry technique for several clinical beams has been studied by Jafari et al. (2014a,b).

These studies to-date focus on the TL properties of variously doped silica. The first of a number of main advantages of using silica beads as an alternative to traditional LiF dosimeters is that the beads are cheap to purchase and can be ordered in bulk quantities. An advantage specific

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to the potential application of *in vivo* dosimetry is that the size and geometry of the beads is such that they can still provide high spatial resolution and additionally glass is chemically inert. The potential application of *in vivo* dosimetry provides motivation for the studies herein, with radiotherapy techniques that utilise beta decay sources as a means of treatment being a particular interest (Morino et al., 2002; Victoria et al., 2013).

The studies herein focus on the potential for doped silica beads used as beta dosimeters. Focusing particularly on *in vivo* dosimetry for instant procedures (Morino et al., 2002) with dose ranges of the order of 10 Gy being delivered. The feasibility of detecting a dose delivered by a beta source in the range of 5 - 20 Gy has been investigated, proposed studies are to extend the dose range to identify the lower limit of detection and investigate the linearity of an increased dose. The range of doses chosen were selected considering doses delivered by the procedure stated in Morino et al. (2002).

2. Materials and Methods

The beads that have been studied for their dosimetric properties were from the manufacturer Mill Hill, Japan. The beads studied herein were, with the exception of the frosted white beads, all transparent, the beads varying in diameter for specific beads; the clear petite, light bronze and dark blue beads being of dimensions 1.58 mm to 1.63 mm and others with diameter of 2.17 mm to 2.20 mm, as displayed in Table 1, with the resolution of the digital caliper being 0.01 mm. The density of the beads has previously been measured to be $2.09 \pm 0.01 \text{ g/cm}^{-3}$ (Jafari et al., 2014a).

2.1. Cleaning

The beads used in this experiment have been used before in previous experiments, cleaned as described by Jafari et al. (2014b), with use of an ultrasonic bath and 10-20 % nitric acid solution.

2.2. Annealing

Prior to annealing beads are strung on to either a stainless steel or aluminium wire, for ease

Bead Type	Photo	Diameter (mm)
White (F)		2.17 - 2.20
Clear Petite (T)		1.58 - 1.63
Clear (T)		2.17 - 2.20
Pink (T)		2.17 - 2.20
Blue (T)		2.17 - 2.20
Red (T)		2.17 - 2.20
Light Bronze (T)		1.58 - 1.63
Dark Blue (T)		1.58 - 1.63
Green (T)		2.17 - 2.20

Table 1: Table of bead properties for each bead type from manufacturer Mill Hill, Japan. Uncertainty in diameter of beads $\pm 0.01\text{mm}$, precision of digital caliper. F and T correspond to the bead either being F-frosted or T-transparent.

of handling. The string of beads were annealed in a Pickstone oven in air (Island Scientific Ltd, Ventnor, UK) equipped with a Eurotherm controller/programmer type 815 by heating to in excess of 400 °C at a rate of 600 °C/hour, then maintained for an hour and left to cool naturally in order to reduce the risk of stress fractures and associated strain-related defect generation.

2.3. Storage

Prior to irradiation and readout, measures need to be taken to reduce the amount of light to which the beads are exposed, to reduce the likelihood of filling superficial traps. The string of beads are wrapped in aluminium foil and stored in a light-tight box, the beads only being taken out for irradiation and subsequent readout of their thermoluminescent response.

2.4. Irradiation

Irradiation of the beads was carried out using the $\text{Sr}^{90}/\text{Y}^{90}$ source contained within the readout device; Risø TL/OSL (see section 2.5). The activity of the source is approximately 37 MBq and has a dose rate characterized for quartz of approximately 0.02 Gy/s. The source emits beta particles at a maximum energies of 0.546 MeV and 2.27 MeV (Dixon et al., 2016). Doses of 5, 10, 15 and 20 Gy were delivered to the beads within the reader one at a time for varying lengths of time (250 s, 500 s, 750 s, 1000 s, respectively) to deliver the desired dose level. The Risø TL/OSL machine has a sample size restriction, specifically in regard to the height of the samples that can be analysed, when located in the samples receptacle, being restricted to a maximum of 2.0 mm. The source is at a distance of 7 mm from the bead when irradiation occurs (DTU Nutech, 2015), reduced to between 5 mm and 6 mm depending on the height of the bead under irradiation. The details of the irradiation for the X-ray irradiation data can be found in Ley et al. (2017).

2.5. Readout

The thermoluminescence readout system used in these experiments is a Risø TL/OSL facility manufactured by the Risø National Laboratory, Technical University of Denmark (DTU). The Risø facility was operated in the preheat setting to acquire data throughout the heating cycle, recording the profile of the glow-curve. The beads were heated in a nitrogen environment, from room temperature to a maximum of 370 °C at a heating rate of 10 °C/s. The planchettes used to hold the samples were stainless steel, produced by the DTU-located manufacturers, DTU Nutech, with the beads placed with the hole axis perpendicular to the planchette surface when readout. Beads were read-out immediately after irradiation unless otherwise stated, such as read-outs conducted in section 3.3.

2.6. Data Analysis

Data is recorded and exported from a program designed for the output of the Risø TL/OSL named Analyst. It is exported into excel where

the glow-curves can be compared and used to identify anomalies in the curves. From the Excel templates a MATLAB code can then be used to sort and subtract background measurements for all glow-curves and to compare yields of different batches of readouts from the Risø TL/OSL. For these studies the background measurements consist of a series of glow-curves following the same heat readout as a sample but instead for an empty planchette, these several glow-curves are averaged and the background for each data point is calculated.

As mentioned in previous work (Ley et al., 2017), the technique used to keep an unbiased comparison between beads with varying magnitudes of signal to noise ratio is to cut off the area of the glow-curve with high contributions to the TL yield due to infra-red radiation. In this work and comparison to previous work the cut off point of the glow-curve is just after the plateau of temperature, this was applied to all coloured beads underestimating the yield of beads producing high TL yields more significantly (for more details see (Ley et al., 2017)).

3. Results and discussion

3.1. Comparison to X-ray Irradiation

The data displayed in this section was collected from the same group of beads that have been discussed in previous work (Ley et al., 2017), with both batches being exposed to 10 Gy but from different sources; a beta source as discussed in section 2.4 and a 6 MV photon beam provided by a Varian electron linac (Varian Medical Systems, Palo Alto, CA).

As shown in Fig. 1, the TL response was greatest from the colourless beads, as found with the X-ray irradiation conducted previously, however the TL yield is reduced when comparing beta to X-ray irradiation by a factor of approximately 10 for all beads (except Green due to difficulty in calculation due to poor TL yield). Using the ESTAR database by Berger et al. (1998) for silicon dioxide the penetration depth for a beta source at the average energy of a $\text{Sr}^{90}/\text{Y}^{90}$ source, 0.2 MeV (Dixon et al., 2016), was estimated to be 0.037 mm. This

Figure 1: Comparative plot of mass normalised average TL yield for Mill Hill, Japan beads irradiated to a dose of 10 Gy by 6 MV LINAC (X-ray source) and $\text{Sr}^{90}/\text{Y}^{90}$ (Beta source), read using the Risø TL/OSL. The error bars were calculated by using the error in the mean.

depth estimate is proposed to be the reason for the X-ray TL yield being tenfold larger than that of the beta response. It is likely that the reduction in response when comparing beta response to X-ray response is due to the geometry of the bead and the distribution of the dose throughout the entire volume of the bead.

3.2. Linearity Study

The study of the TL response for different coloured beads with increasing dose showed a strong linear correlation for all coloured beads, except green, as shown in Figs. 2 & 3. There is a strong linearity for the clear, white, clear petite and pink beads shown for doses 5 - 20 Gy, as shown by Fig. 2, with best fit lines calculating a linearity gradient of 4556.1, 5400.0, 5586.4, 779.7 counts $\text{mg}^{-1}\text{Gy}^{-1}$, with errors displayed in Table 2, and R^2 values of 0.9973, 0.9974, 0.9998, 0.9950, respectively.

The dose response of the remaining colours of silica beads is displayed in Fig. 3, showing comparatively lower mass normalised TL yields to the pink and colourless beads, however almost all beads show linearity at this dose range for beta irradiation with the exception of green beads. The blue, red, light bronze and dark blue beads show a linear response to an increase in the dose received and display reasonable R^2 values to the lines fitted, displayed in Table 2. The green beads do not

show the linear response as they produce a poor TL response at 5, 10 and 15 Gy with a low signal to noise ratio; the data point at 20 Gy has a large uncertainty in the value due to the difficulty in determining TL signal from raw data. It is suggested that the poor TL yield acquired from the green coloured beads is related to self-attenuation.

Bead	Gradient of Linear Fit (counts $\text{mg}^{-1}\text{Gy}^{-1}$)	Error in gradient (%)	R^2 Value
Clear Petite	5586.4	7.6	0.9998
White	5400.0	11.0	0.9974
Clear	4556.1	11.2	0.9973
Pink	779.7	5.9	0.9950
Blue	166.7	3.5	0.9976
Red	121.7	5.0	0.9950
Light Bronze	62.2	6.5	0.9914
Dark Blue	26.9	18.9	0.9332
Green	No linear fit due to poor TL yield		

Table 2: Gradient, error in gradient and R^2 values of best fit lines fitted to the data of the different beads as displayed in Figs. 2 & 3. The error in the gradient represents the percentage of the standard error.

The y-intercepts of the trend lines fitted to the data in Figs. 2 & 3 range between 173.5 and -9741.1, with all of the beads showing a high TL yield per mg with an interception on the axis of TL yield less than zero, this could be evidence for a lower detection limit for TL response to beta decay in these beads, but more directly a result of statistical uncertainty at low dose.

The results from all coloured beads, except green, demonstrate that the silica beads have potential to be used for applications of treatment using beta emitters as dosimeters, particularly for such treatment techniques as in-stent restenosis (Morino et al., 2002).

3.3. Fading Study

The fading of the TL response from the clear petite beads was studied by varying the time be-

Figure 2: Graph displaying linear dose response for beads, irradiated with $\text{Sr}^{90}/\text{Y}^{90}\beta$ source, producing comparatively high TL yield, readout using the Risø TL/OSL. Error bars calculated by using the error in the mean. Corresponding equations for the trend lines and R^2 values are displayed in the legend.

Figure 3: Graph displaying linear dose response for beads, irradiated with $\text{Sr}^{90}/\text{Y}^{90}\beta$ source, producing comparatively low TL yield, readout using the Risø TL/OSL. The solid line connecting the green data points is to aid in identifying them and is not presented as a line of fitting the data. Error bars calculated by using the error in the mean.

Figure 4: Typical glow-curves of a clear petite bead irradiated with a dose of 5 Gy with increasing time between irradiation and read-out, indicated by the legend (in minutes), with the black dashed line showing the heat-cycle of the read-out.

Figure 5: Typical glow-curves of a clear petite bead irradiated with a dose of 20 Gy with increasing time between irradiation and read-out, indicated by the legend (in minutes), with the black dashed line showing the heat-cycle of the read-out.

tween irradiation and read-out, relevant to the time scale of the applications considered herein. During the delay in read-out, light-tight containment of the beads was provided by retaining them in the Risø reader at room temperature. For the two extremes of dose investigated, the beads were read out following a short delay time (the order of two minutes) post-irradiation (excluding the quench time of the detector as necessary in an applications procedure of readout) through to up to 70 hours. The nature of the fading can be seen from Figs. 4 & 5, displaying glow-curves of a particular bead irradiated with 5 Gy and 20 Gy, respectively, for different time periods between irradiation and readout. Figs. 4 & 5 both show a sequential pattern between the shape of the glow-curve and the time interval between irradiation and read-out; with the shortest time interval between irradiation and read-out shown as the outer glow-curve (largest peak) and the longer time intervals shown as the inner glow-curves (smallest peaks).

The effect the fading had on the TL response of the beads for each dose, on average, is displayed in Fig. 6. The errors in the TL yield per mg were taken as the error in the mean of the TL yield per mg response of each set of samples, the errors being found to be between 1.04 % and 2.15 %. This error is suggested to be largely representative of sample inhomogeneity across the range of beads used.

It was found that the beads showed a fading of approximately 10 % of the TL yield per mg when comparing the immediate read-out and the 1 hour read-out. These fluctuations appear to differ by comparable percentage errors in the mean of the samples after 18 hours between irradiation and read-out. The data displays a plateau in the percentage of fading after delaying the read-out by one day after irradiation, as shown by Fig. 7, this plateau occurring at approximately 17 % and 22 % loss of signal for 5 Gy and 20 Gy doses respectively. The errors displayed in Fig. 7 were calculated by taking the percentages of each bead in each of the samples and then averaging across the percentage of the immediate signal measured in comparison. These errors were measured to be

Figure 6: Mass normalised TL yield, for both 5 Gy and 20 Gy, for increasing time between irradiation and read-out. Errors calculated by the error in the mean of the samples.

≤ 1.3 %. The data of the 5 Gy and 20 Gy TL yield/mg is fitted to a logarithmic trend with R^2 values of 0.9883 and 0.9705 respectively.

A further study for 30 minute fading read-out was repeated with varying times between annealing and irradiation, to investigate the reproducibility of the TL response for individual beads. The majority of beads showed a reproducibility on average of 0.54 % of the average of the two TL responses, the largest being 1.01 %, (with the exception of a bead showing a 6 % reproducibility due to abnormally large background signal).

The widely observed fading phenomenon that occurs over the period of storage at ambient temperature is a thermally-driven cascading (relaxation) process among the electron traps. It is likely that the severity of the fading varies with bead colour, this would be an area for further study on the beads showing higher TL responses for beta irradiation.

4. Conclusions

This study has demonstrated that doped silica beads have the potential to be used for beta source dosimetry with dose rates as low as 0.02 Gy/s. When comparing to an X-ray source the mass normalised TL yield is smaller by an order of magnitude, most likely due to the LET of the beta particles when interacting with the silica bead.

Figure 7: The average percentage of TL yield as a function of time (minutes) compared to the immediate read-out. Errors calculated by taking the percentages of each bead in each sample and then averaging across the fading percentage of immediate TL response and relative fading TL response.

Also demonstrated is that all bead colours, except green, manufactured by Mill Hill, Japan, show a linear response to an increase in dose. Certain coloured beads show a suitable TL yield and linear response to increasing beta irradiation in the dose range typical of some radiotherapy procedures (Morino et al., 2002), with; white, clear, clear petite and pink beads being the most suited and the clear petite bead showing the strongest conformity to a linear correlation with an R^2 value of 0.9998 and producing the highest mass normalised TL yield.

The fading study has shown that the TL response is mainly affected by the fading phenomenon within the first hour between irradiation and read-out. Although small fluctuations do still occur this is most likely due to the homogeneity across the batch of samples used. The mass normalised TL yield flattens out at approximately 17 % and 22 % loss of signal for 5 Gy and 20 Gy doses respectively. Further studies into the reproducibility of beads showed that the mass normalised TL yield varied by 0.54 % on average. It is likely that the reproducibility can be further improved upon by maintaining the same

time between annealing and irradiation, prospects for which could be undertaken as further study as well as the affect fading has on the TL response of different coloured beads.

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References

- Abdul Sani, S.F., Alalawi, A.I., Hairul Azhar, A.R., Mahdiraji, G.A., Tamchek, N., Nisbet, A., Maah, M.J., Bradley, D.A., *High sensitivity flat SiO₂ fibres for medical dosimetry* Radiation Physics and Chemistry 104, 134-138, 2014.
- Bradley, D.A., Abdul Sani, S.F., Alalawi, A.I., Jafari, S.M., Noor, N.M., Azhar, A.R.H., Mahdiraji, G.A., Tamchek, N., Ghosh, S., Paul, M.C., Alzimami, K.S., Nisbet, A., Maah, M.J., *Development of tailor-made*

- silica fibres for TL dosimetry* Radiation Physics and Chemistry 104, 3-9, 2014.
- Dixon, J., Rajan, A., Bohlemann, S., Coso, D., Upadhyaya, A. D., Rohatgi, A., Chu, S., Majumdar, A., Yee, S., *Evaluation of a Silicon ^{90}Sr betavoltaic power source*, Scientific Reports 6, Article number: 38182, doi:10.1038/srep38182, 2016.
- DTU Nutech, *Guide to The Riso TL/OSL Reader*, DTU Nutech, Denmark, August, 2015.
- Berger, M. J., Coursey, J.S., Zucker, M.A., Chang, J., *Stopping-power & range tables for electrons, protons, and Helium ions*, NISTIR 4999, 1998 - Last update: July 2017, https://physics.nist.gov/cgi-bin/Star/e_table.pl.
- Jafari, S., Bradley, D.A., Gouldstone, C.A., Sharpe, P.H.G., Alalawi, A., Jordan, T.J., Clark, C.H., Nisbet, A., Spyrou, N.M., *Low-cost commercial glass beads as dosimeters in radiotherapy*, Radiation Physics and Chemistry 97, 95-101, 2014.
- Jafari, S., Jordan, T.J., Hussein, M., Bradley, D.A., Clark, C.H., Nisbet, A., Spyrou, N.M., *Energy response of glass bead TLDs irradiated with radiation therapy beams*, Applied Radiation and Isotopes 104, 208-211, November, 2014.
- Ley, K., Jafari, S., Lohstroh, A., Bradley, D. A., *Investigations of thermoluminescent silica beads of different manufacturers and colours*, Submitted to Radiation Physics and Chemistry.
- Mizanur Rahman, A. K. M., Zubair, H. T.; Begum, Mahfuza; Abdul-Rashid, H. A.; Yusoff, Z.; Ung, N. M.; Mat-Sharif, K. A.; Wan Abdullah, W. S.; Amouzad Mahdiraji, Ghafour; Amin, Y. M.; Maah, M. J.; Bradley, D. A., *Ge-doped silica optical fibres as RL/OSL dosimeters for radiotherapy dosimetry*, Radiation Physics and Chemistry 116, 170-175 July, 2015.
- Morino Y., Kaneda H., Fox T., Takagi A., Hassan A.H.M, Bonan R., Crocker I., Lansky A.J., Laskey W.K., Suntharalingham M., Bonneau H.N., Yock P.G., Honda Y., Fitzgerald P.J., *Delivered dose and vascular response after β -radiation for in-stent restenosis: retrospective dosimetry and volumetric intravascular ultrasound analysis*, Circulation 106, 18, October 29, 2002, <https://doi.org/10.1161/01.CIR.0000036367.17043.03>.
- O'Keeffe, S., Woulfe, P., Sullivan, F.J., *Radioluminescence based optical fibre sensor for radiation monitoring during brachytherapy*, Proceedings of IEEE Sensors (2015), DOI 10.1109/ICSENS.2015.7370523, January 2016.
- Victoria M. L. Cohen, Vasilios P. Papastefanou, S. Liu, Ian Stoker, and John L. Hungerford, *The use of Strontium-90 beta radiotherapy as adjuvant treatment for Conjunctival Melanoma*, Journal of Oncology, 2013, Article ID 349162, 7 , doi:10.1155/2013/349162.