Ge-doped silica optical fibres as RL/OSL dosimeters for radiotherapy dosimetry

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ABSTRACT

Various tailor-made doped-silica optical fibres are investigated as dosimeters in support of radiotherapy, based on radioluminescence (RL) and optically stimulated luminescence (OSL) technology. Investigations focus on the development of these glassy dosimetric media, offering a number of advantages going well beyond their water impervious nature and excellent spatial resolution (~few microns). An RL/OSL photomultiplier-tube (PMT)-based reader was assembled, providing for study of the influence on the RL/OSL signal of different Ge-dopant concentrations (3.59, 4.74 and 7.03 wt%) in silica fibres exposed to medical LINAC photon beams. Among the three arbitrary choices of dopant concentration, those fibres containing the least concentration of Ge (3.59 wt%), denoted as Ge-1, gave rise to the greatest RL yield, at 1.67 and 2.34 times that of Ge-2 (fibres Ge-doped at 4.74 wt%) and Ge-3 (fibres Ge-doped at 7.03 wt%) respectively, reducing in yield with increasing Ge-dopant concentration. At 7.3 × 10³ counts Gy⁻¹ min at 22 °C, the Ge-1 fibres provided the superior sensitivity, also being found to be reusable without noticeable variation in RL signal (<1%; 1 SD) for X-ray exposures delivered at a dose-rate of 600 cGy/min. The RL signal was found to be free from spectral superposition or noise, also exhibiting energy independence in the use of X-rays generated at 6- and 10 MV. In regard to percentage depth-dose (PDD), in measurements made using optical fibre dosimeters and 6 MV X-ray photon beams, the maximum value of PDD, dmax, was obtained at a depth of 1.5 cm, in accord with ionization chamber measurements. Using green stimulation light, for all three concentrations of Ge-dopant, linearity between the OSL signal and dose was observed across the 0.5- to 8 Gy dose range investigated. Results from this ongoing study are intended to assist in efforts towards improving the performance of RL/OSL fibre sensors for radiotherapy dosimetry.

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1. Introduction

The 2014 World Cancer Report of the International Agency for Research on Cancer (IARC) made the estimate that annual deaths from cancer would rise from 8.3 million in 2012 to 13 million by 2034 [1]. Radiotherapy, an effective treatment option, has been identified as one of the possible therapeutic solutions. Advanced cancer treatment modalities such as volumetric modulated arc radiotherapy (VMRT) and intensity modulated radiotherapy (IMRT) have made highly conformal treatment plans possible, offering reduced risks to healthy tissues. The reliability of dose verification for these complex clinical routines requires real-time in-vivo dosimetry [2], in recent years radioluminescence (RL) dosimetry becoming of increasingly popular use in such circumstances [3]. However, when the dosimetric probe is placed in a radiation field an unavoidable consequence is that a portion of signal carrier fibre is also exposed, producing an additional contribution to the overall signal, a situation commonly referred to as the “stem effect”. This contribution is mainly due to Cerenkov radiation, as well as fluorescence or luminescence light emission, superimposed onto...
the RL signal from the dosimetric sensor [4,5]. In addition to the real-time RL dosimetry technique, the possibility also arises that optically stimulated luminescence (OSL) can be harnessed to provide “post irradiation” readout, representing an independent signal for dose estimation. The basis of OSL measurement is to stimulate a pre-irradiated sample with an appropriate wavelength of light and to monitor the consequent emission from the sample at a different wavelength [6,7].

To detect ionizing radiation, further acting as dosimeters, use is currently being made of a number of luminescence-based devices, one form of which is the focus of present interest. Such devices can be in the form of solids, liquids or gases. Metal-oxide-semiconductor field-effect transistors (MOSFETs), p-type diodes, ionization chambers, diamond detectors, TLDs, film (radiographic and radiochromic) and the chemically-based Fricke detector system have all been adopted as reference-, relative-, on-line, active- and passive-dosimeters (see for instance, Izewska and Rajan [8]). Active dosimeters are typically electronic devices, providing direct (on-line) dose evaluation, whereas the passive dosimeters (which include thermoluminesence, optically stimulated luminescence, and numerous other diverse forms) store the irradiation information (a form of non-permanent radiation damage), each giving subsequent dose information through use of an off-line form of readout. Each category and form of device offers advantages and disadvantages in accord with the particular applications, not least in respect of expense and convenience.

For radiotherapy, in seeking to approximate the performance of an ideal dosimeter, such devices should be small in size (down to sub mm) yet highly sensitive, also offering large dynamic range (a wide range of response to dose, from mGy to the order of some 10 s of Gy). Among the above-mentioned sensors, none meet all such requirements [9]. In recent times silica-based optical fibre dosimeters have gained popularity as radiation dosimeters, due in no small part to their excellent spatial resolution (with dimensions of a few tens of microns), water insolubility, chemical inertness, free of risk from various hazards (fire, explosions, electromagnetic interferences) and affordability. Such optical fibres typically consist of an optically transparent doped core surrounded by a transparent cladding material. In traditional application for use in communications, to ensure total internal reflection the refractive index of the core of the fibre is made greater than that of the cladding by the addition of the dopant. It is with respect to such construction, namely of a doped core insulator, that as a matter of serendipity, the selectively doped silica fibre core also forms the basis of a passive radiation dosimeter (see for instance [10]). The dosimetric properties of SiO2 optical fibres depend on the trapping processes caused by the occurrence of structural defects in the material [11].

The radiation sensitivity (hampering) of telecommunication fibres has now been known for many years. Specifically, for optical fibres exposed to ionizing radiation, it is known that the data transmission capability of the fibre can be affected (attenuated) through radiation induced ionization and formation of traps (colour centres), created by the presence of impurities within the optical fibre core [12,13]. More recently it has been realized that the presence of the colour centres can be correlated with the radiation dose. By using the associated mechanism of radiation hammering, optical fibres can be evaluated for their utility as radiation detectors rather than in study of the deterioration of their optical communication capability. Three intimately related luminescence phenomena, RL, OSL and thermoluminescence (TL), all based on the energy band structure of materials, can be independently harnessed for use in radiation dosimetry. RL is the spontaneous fluorescence emitted from scintillating material by recombination of the electron-hole pairs caused by the immediate irradiation, also being directly associated with the dose rate. In an RL type dosimetry system the active portion of dosimeter can be made for instance of a small (sub mm2) piece of scintillator optically attached to the tip of a long polymethyl methacrylate (PMMA) fibre cable. When the sensor is exposed to ionizing radiation, an optical (light) signal is generated and is guided through the PMMA fibre towards a detecting device placed at a distance (metres and more) from the radiation zone. RL type sensors provide real-time information. Conversely, pre-existing traps, populated for instance as a result of absorbed dose, can lead to luminescence emission upon receipt of additional energy from an external source of stimulation of the appropriate wavelength of light (generally in the visible range), termed OSL, or by heat stimulation, termed TL, both with intensity related to the amount of absorbed dose.

The luminescence properties of Ge-doped SiO2 fibre are affected by dopant concentration along the core of the fibre. Owing to the presence of Ge-atoms (impurities) in the silica glass matrix, colour centre formation is facilitated: since the energy of the Ge-0 bond (~3.6 eV) is lower than that of the Si-0 bond (~5 eV), the rupture of these bonds by incident radiation is more probable than that of the Si-0 association [14]. The details of both the nature and the molecular structure of radiation-induced point defects in pure and doped glassy silica has been described elsewhere [15,16].

Present dosimetric study focuses on the RL and OSL characteristics of lab-fabricated Ge-doped fibres of core diameter 100 μm, manufactured using the Modified Chemical Vapour Deposition (MCVD) process. In seeking to fulfill the requirements of a sensitive form of dosimetry there is need to investigate dependency on the quantity of Ge-dopant, a situation that is not usually to be attained through use of commercially available telecommunication optical fibres, these generally being of a particular dopant concentration. The particular interest is investigation of the effect of doping concentration of SiO2 glass based on the RL/OSL method, pointing to optimum Ge-dopant concentrations for sensitive radiotherapy dosimetry. This will depend on source types, in present study these being penetrating (energetic, ~MeV) X-ray photons.

2. Materials and methods

2.1. Dosimeter fibre fabrication and characterization its quality standards

Three different Ge concentrations were selected in the doping of silica optical fibres, guided by the relatively limited amount of a priori information available on luminescence yield for such systems. Here it should be mentioned that several of the research team within this group have also been involved in the investigation of the TL properties of different Ge-concentrations in silica optical fibres [17,18], it being known from those studies that the TL yield for Ge-doped silica are close to optimum at around the value choices that have been retained herein. Doping was through use of the MCVD method, with details of the fabrication procedure also being available elsewhere [19,20]. The fibre fabrication process consists of two steps: preform fabrication wherein the doping takes place and the drawing process to produce fibres. The preforms were pulled into fibre with core/cladding diameters (100/604) μm. It is expected that larger cross-sectional fibre samples will provide greater number of traps [21], cross-sections that would need to be compatible with the larger core-sized PMMA fibre used herein.

In previous studies, SEM (Scanning Electron Microscope)-based EDX (Energy Dispersive X-ray Spectroscopy) analyses have been used in X-ray mapping and line-scan profiling, performed to determine the relative presence of doping material in the silica fibre core. With use of an optical cleaver, the fabricated optical fibres were cut into 1 cm lengths for SEM-EDX analysis. Ease of incident electron flow to the ground was ensured through use of carbon tape attached to the sample, also the low atomic number of carbon ensuring min-
Table 1

<table>
<thead>
<tr>
<th>Sample identifier</th>
<th>EDX analysis</th>
<th>Effective atomic number ($Z_{eff}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ge-1</td>
<td>O 59.0</td>
<td>13.4</td>
</tr>
<tr>
<td></td>
<td>Si 37.4</td>
<td>26.2</td>
</tr>
<tr>
<td></td>
<td>Ge 3.59</td>
<td>0.98</td>
</tr>
<tr>
<td>Ge-2</td>
<td>O 57.6</td>
<td>14.1</td>
</tr>
<tr>
<td></td>
<td>Si 37.7</td>
<td>26.8</td>
</tr>
<tr>
<td></td>
<td>Ge 4.74</td>
<td>1.31</td>
</tr>
<tr>
<td>Ge-3</td>
<td>O 54.8</td>
<td>14.9</td>
</tr>
<tr>
<td></td>
<td>Si 38.2</td>
<td>27.9</td>
</tr>
<tr>
<td></td>
<td>Ge 7.03</td>
<td>1.9</td>
</tr>
</tbody>
</table>

Fig. 1. Individual reconstructed images of Ge (left) and Si (right) of three different concentration Ge-doped fibre.

Table 1: Element composition of the different dopant concentration doped SiO$_2$ optical fibres.

Table 1 presents mapping analysis results for distribution of Ge-dopant in the core (left-hand panels) and SiO$_2$ distribution (right-hand panels). Increase in the concentration of Ge provides for a more readily apparent relative presence of the dopant in the silica matrix.

2.2. Probe preparation with the Ge-doped silica optical fibre dosimeter

The RL/OSL sensor probes were prepared from the fabricated Ge-doped optical fibre, each of 2 cm length, coupled with the core of $\sim$1 mm PMMA optical fibre (SH4001 Super ESKA, Mitsubishi, Japan) of length 10 m. These samples were inserted into a polyoxymethylene (Delrin) ferrule and one end was aligned with the core of polished PMMA fibre. The in-house designed probe holder
2.4. Experimental procedures

In present work, the RL/OSL response was examined using 6- and 10 MV X-ray exposures produced by a medical linear accelerator (LINAC, Varian 2100C/D). The source-to-surface (SSD) distance was set at 1 m and the irradiation field size to 10 × 10 cm². The probe-head (the laboratory-fabricated Ge-doped optical fibre) was positioned at dose maximum, d max (1.5- and 2 cm below the phantom surface for 6- and 10 MV beams respectively) using a solid-water™ (Gammex, USA) phantom. The RL/OSL signal was carried by means of a 10 m length of PMMA fibre through a thick wall offering radiation protection, linking to the acquisition terminal outside of the radiation chamber. Fig. 6 shows the actual hospital-environment measurement set-up.

Two PMMA fibre cables, one with the Ge-doped fibre sample and another without, were irradiated under identical circumstances. The background signal from stem effects (resulting from Cerenkov light and fluorescence) and inherent RL from the PMMA fibre was recorded with the bare PMMA fibre cable, then be subtracted from the acquired total photon counts recorded using the dosimeter sample (RL from dosimeter + RL from PMMA fibre). The resultant RL yields were then used in evaluating the dose-rate, linearity of response to dose, energy dependence and percentage depth dose (PDD).

2.5. Data acquisition and analysis of the luminescence signals

The RL measurements were carried out using a 1 Hz sampling rate. Irradiation against dose rates from 100- to 600 Gy/min was carried out using a 100 Gy/min step. The RL signal was analysed for dose-rate response, dose response linearity, energy dependence and percentage depth dose (PDD), while for OSL measurements the Ge-doped fibre samples were irradiated to doses ranging from 0.5- to 8 Gy. The OSL signals were captured using a 2 Hz sampling rate stimulated by a 30 mW green LED light (stimulation time 800 ms). The dose response was obtained by integrating the OSL signal over the irradiation time.

3. Results and discussion

3.1. RL characteristics of Ge-doped silica optical fibre

For effective real-time RL dosimetry in a clinical environment, several demands must be satisfied, including reproducibility, linearity to absorbed dose and independence from dose-rate. Additionally, dose measurements using the fibre-coupled RL dosimeter should be free of parasitic stem signals generated in the signal carrier, PMMA optical fibre in present work. For the three forms of Ge-doped optical fibre (Ge-1, Ge-2 and Ge-3), Fig. 7 shows the background-corrected RL response, obtained at a dose-rate of 600 Gy/min using 6 MV X-ray photon irradiation and an exposure time of 50 s. Over the duration of exposure the RL signal was observed to remain constant; also observed is that the RL intensity decreased with increase in Ge-dopant concentration, Ge-1 (3.59 wt%) providing the greatest response of the three different Ge-concentration fibres. Of note in this respect is that concentration quenching can result from strong non-radiative recombination (retrapping) in the amorphous medium. Hence, while a greater number of defect centres might be expected to be associated with greater light yield, an enhancement of Ge-doping can also be expected to enhance interaction between defect centres, also
more greatly disturbing local periodicity \cite{23,24}. Thus taking into account the effect of concentration quenching, including retrapping, further increase in doping beyond an optimal value can be expected to limit rather than enhance RL light yield.

Fig. 8 shows the absorbed dose versus RL response for the three concentrations of Ge-doped optical fibre, subjected herein to 6 MV X-ray photon irradiation, delivering doses in the range 0.01- to 10 Gy. The RL responses for all three fibre types are seen to increase with dose, the lowest Ge-concentration optical-fibre (Ge-1) producing the greatest RL response, with total photon counts some 1.65 and 2.34 times that of the Ge-2 and Ge-3 fibres respectively. Also apparent is that all three fibres provide excellent linearity of RL against dose (also see Figs. 9–11 below). Implicit in regard to the slope values, all rather steep (i.e., highly sensitive to dose-rate, as desired), is that with such steepness small change in slope (say 5%) can reflect in a large change in intercept. The reality of such an outcome is that each and every probe that is intended for dosimetric application would need to be individually calibrated.

Energy dependence investigations of photons generated at 6- and 10 MV and dose-rates from 100- to 600 cGy/min, concern the three types of Ge-doped fibre sensor placed at d_{max}, with SSD = 100 cm and a 20 s data acquisition gate time. In each case, RL increases linearly with dose-rate, responses separating marginally at higher dose-rate (Figs. 9–11), attributed to the differing deep trap
distribution in the host glass matrix [25]. Apparent is the importance of evaluating energy absorption variation in the material with radiation energy.

For a 10 × 10 cm² field size and a 600 cGy/min dose rate, 6 MV X-ray depth-dose measurements have been carried out using the Ge-doped RL sensors at various depths 0- to 16.5 cm in solid water™. The results of Fig. 12 show there to be agreement among the doped fibres, although for depths beyond 3 cm fibre values are larger than that of the ionization chamber (using a CC13 ion chamber of volume 0.13 cm³; IBA, USA). This may be attributable to residual stem effects within the signal-carrying PMMA fibre (most particularly as a result of Cerenkov light generation), a result of what may well be the use of a potentially imperfectly matched pair of PMMA fibres applied in backing-off the stem effect (the reader is referred back to Section 2.4 for a discussion of this). Subsequent investigations, for dosimeters exposed to larger field sizes and consequently with larger portions of the PMMA fibre irradiated, have shown increased discrepancy due to Cerenkov radiation, account being taken of the difference in effective atomic number of the fibre and water.

Radiation output intensity fluctuations are common in almost all types of linear accelerator owing to beam stabilization and pulse-to-pulse variation. Variation in dose-rate is typical at the initiation of an exposure, stabilizing over several seconds. Fluctuations in LINAC output should be well within manufacturer specifications and have not been shown to affect the total dose delivered during radiotherapy treatment [26].

For the 6 MV X-ray irradiations of Fig. 13(a), the dose-rate levels show minimal initial fluctuation in approaching stabilization. Conversely, at the initiation of each 10 MV X-ray irradiation exposure, for each dose-rate level, much larger initial fluctuation is observed, the level of initial peak height in each case increasing to a saturation value with increase in dose-rate (from 100 cGy/min...
Fig. 9. Megavoltage X-ray response of Ge-1 fibres, showing marginal energy dependence. See the caption of Fig. 8 for comments on uncertainties in slope and intercept.

Fig. 10. Megavoltage X-ray response of Ge-2 fibres at various dose-rates. See the caption of Fig. 8 for comments on uncertainties in slope and intercept.

Fig. 11. Megavoltage X-ray response of Ge-3 fibres at various dose-rate. See the caption of Fig. 8 for comments on uncertainties in slope and intercept.

Fig. 12. Comparison of 6 MV X-ray PDD using the Ge-doped fibres and a standard ionization chamber.

Fig. 13. Use of the Ge-doped RL system to monitor the stability of delivered dose-rates from a Varian 2100C/D LINAC, X-rays being generated at different potentials (6- and 10 MV).

Fig. 14 shows the normalized RL (normalized to 600 cGy/min) (Fig. 13(b)). At this beam energy, for the UMMC LINAC the release dose-rate at each level is seen to be greater than the specified dose-rate, subsequently stabilizing to the design level over a period of a few seconds as previously mentioned. Most apparent in using this Ge-doped RL dosimetry system is that true real-time monitoring of dose-rate is possible, offering direct feedback to the operator.

For all three types of Ge-doped fibres, use being made of the 6 MV beam delivered at a dose-rate of 600 cGy/min, comparison has been made of stability of response resulting from repeat exposure of the fibres. RL emissions have been recorded over ten consecutive irradiation cycles, the exposure time being 30 s, with a 60 s lapse between irradiations. Fig. 14 shows the normalized RL (normalized
to mean values) as a function of cycle number. Apparent is that the RL response shows no practical change between the consecutive measurements, relative readings being well within ±0.02%, with a standard deviation of 0.012% obtained for the series of 10 readings. The small variation in RL results is random, offering excellent reproducibility of the RL signal.

3.2. OSL characteristics of Ge-doped silica optical fibre

OSL investigations were conducted using the three different Ge-dopant concentration optical fibres, each of the same dimensions (2 cm length, 604 μm diameter) and of the same mass as those used in the RL dosimetric studies. A typical set of OSL curves for the three different concentrations of Ge-doped optical fibre samples (Ge-1, Ge-2 and Ge-3) are shown in Fig. 15, demonstrating the intensity of the OSL signal to be comparable and also considerably greater than the background of non-irradiated (blank) samples under green-light stimulation. The variation in sensitivity (with reference to initial intensity) with concentration is not well differentiated. The exponential nature of signal decay of the SiO₂ optical fibres, as previously reported in the literature [27], is re-obtained in present results. It is to be noted that the stimulant light used herein has yet to be optimized for the optical fibre OSL samples. Also of note, as again demonstrated herein, are that the OSL curves are typically very short-lived under the influence of high intensity light, rapid decay being desired in order to provide fast readout.

The optical fibre sample Ge-1 (3.59 wt% Ge) has been used herein to investigate dependence of dose response on OSL emission (again for the important radiotherapy dose range, 0.5- to 8.0 Gy), as illustrated in Fig. 16. Read-out was performed at room temperature using green light stimulation, supplied from an LED source (515 nm, 30 mW) focused onto the PMMA fibre that has been used to couple to the dosimeter probes. As seen from the graph, OSL decay is more rapid with increase in dose, OSL intensity also increasing with dose.

Only limited literature has been found in regard to the use of commercial optical fibres as passive OSL dosimeters [27,28]. The study of Espinosa et al. was performed under green excitation light, at elevated temperature (~125 °C) rather than at room temperature. For in-vivo fibre-optic-coupled dosimeter (FOCD) radiotherapy dosimetry, in seeking to avoid optical distortion in use of Mitsubishi SH 4001 PMMA fibre as a wave-guiding medium, the manufacturer specifies operating temperatures in the range –55 °C–70 °C. Present study was thus conducted at room temperature, albeit finding the typical set of continuous wave OSL (CW-OSL) decay curves to be analogous to the reported results in the literature [27,28].

The relationship between total luminescence and radiation dose was found to be linear, as shown in Fig. 17. Using the same experimental conditions as above, doses from 0.5 to 8.0 Gy were applied, with a dose rate of the 6 MV X-ray beam of 600 cGy/min, irradiation measurements being made at the dₘₐₓ position in the solid-water™ phantom. Several parameters such as integrated photon counts were calculated, measured in the example case as the area under each curve for particular choice of read-out time (45 s herein). Five
samples were measured at each dose. Plotted as a function of radiation dose, the means and standard deviations for each measured point are as shown in the figure, the OSL response being observed to be linear against dose.

In general, OSL signal decay at any given time is related to the rate at which the charge carrier materials are excited from defect traps to then experience recombination. The integral of the luminescence-versus-time curve is associated with trapped charge concentration, which in turn is proportional to the absorbed dose, the basis of OSL dosimetry.

4. Conclusions

Present Ge-doped silica optical fibres have been confirmed to possess RL/OSL properties that make them suitable for radiotherapy dosimetry. OSL decay curves and RL response linearity and consistency being analyzed over various appropriate dose-rates. The systematic over-estimate of PDD as measured by the fibres were observed to be due to the stem signal noise within the signal-carrying PMMA fibre, arising from Cerenkov radiation, larger field sizes, producing larger contributions from the Cerenkov radiation as expected. Over prolonged periods the recorded RL response was consistent, making the doped silica fibre suitable for real-time dose measurements. Other properties such as high melting point, small dimension (allowing for unsurpassed spatial resolution), large core size (ensuring maximum light propagation) and insolubility in water make this material a promising candidate for radiotherapy dosimetry, both externally and in vivo. In conclusion the doped silica optical fibre sensor, with efficient coupling, can be a viable solution for a range of radiotherapy dosimetry applications as well as a promising choice for reference dosimetry and as a quality assurance tool in advanced radiotherapy developments that are presently coming on-line world-wide.

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References

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Ung Ngie Min is a senior lecturer and medical physicist at the Department of Clinical Oncology, University of Malaya Medical Centre. He possesses Bachelor of Biomed- ical Engineering and Master of Medical Physics from the University of Malaya. He then completed his PhD at the University of Western Australia in collaboration with Genesis Cancer Care (formerly known as Perth Radiation Oncology). His Ph.D. work revolved around the investigation of uncertainties in fiducial markers tracking during image-guided radiotherapy (IGRT) of prostate cancer. Apart from IGRT, his other current research interests include radiation dosimetry, brachytherapy and advanced radiotherapy techniques.

David Bradley, Professor in Radiation Physics, has 40 years of experience in academia, industrial and medical sec- tors. Previously Secretary of the International Radiation Physics Society (IRPS), Editor-in-Chief of ‘Applied Radia- tion and Isotopes’, ‘Radiation Physics and Chemistry’ and the ‘British Journal of Radiology’, he is the author of some 340 publications, with presentations at more than 100 conferences. His research on the fundamentals and appli- cations of photon scattering, radiation dosimetry, radio analytical techniques for determination of trace element concentrations, radiological risks associated with Nat- urally Occurring Radioactive Material (NORM) and the development of silica-based luminescence dosimeters is well-documented.