High sensitivity flat SiO₂ fibres for medical dosimetry

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HIGHLIGHTS

- Studies of a novel undoped flat fibre fabricated for medical dosimetry.
- Investigation of the TL response of the undoped flat fibres, TLD-100 chips and Ge-doped fibres in megavoltage potentials.
- The sensitivity of the flat fibres is found to be markedly higher than that of Ge-doped fibre, being comparable to the TLD-100.

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ABSTRACT

We describe investigation of a novel undoped flat fibre fabricated for medical radiation dosimetry. Using high energy X-ray beams generated at a potential of 6 MV, comparison has been made of the TL yield of silica flat fibres, TLD-100 chips and Ge-doped silica fibres. The flat fibres provide competitive TL yield to that of TLD-100 chips, being some 100 times that of the Ge-doped fibres. Pt-coated flat fibres have then been used to increase photoelectron production and hence local dose deposition, obtaining significant increase in dose sensitivity over that of undoped flat fibres. Using 250 kVp X-ray beams, the TL yield reveals a progressive linear increase in dose for Pt thicknesses from 20 nm up to 80 nm. The dose enhancement factor (DEF) of $(0.0150 \pm 0.0003) \text{ nm}^{-1} \text{ Pt}$ is comparable to that obtained using gold, agreeing at the 1% level with the value expected on the basis of photoelectron generation. Finally, X-ray photoelectron spectroscopy (XPS) has been employed to characterize the surface oxidation state of the fibre medium. The charge state of Si2p was found to lie on 103.86 eV of binding energy and the atomic percentage obtained from the XPS analysis is 22.41%.

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1. Introduction

Dosimetry is fundamental to many applications of radiations, with numerous investigations made of a wide range of dosimeter forms, including radiation-induced attenuation in optical fibres (Huston et al., 2001), radioluminescence (RL) and optically stimulated luminescence (OSL) (Justus et al., 1997; Aznar et al., 2004; Benevides et al., 2007). Thermoluminescence dosimetry (TLD) is well-established, often considered to be the most widely used and

cost-effective approach to radiation dosimetry, not least in regard to radiation therapy (Hashim et al., 2006).

While phosphor TLD media such as LiF have been widely exploited in medical dosimetry, these have several acknowledged defects, including potential hygroscopic issues and relatively poor (~mm) spatial resolution (Izewska and Rajan 2005). In recent years, a number of investigations have concerned the TL properties of doped silica (SiO₂) optical fibres, overcoming the particular limitations of existing TLD systems (Hashim et al., 2009), pointing to intercalary and interstitial measurement applications and high sensitivity over a wide range of dose rates and absorbed doses (Yusoff et al., 2005). The TL response of commercial doped SiO₂ optical fibres has been investigated for photons (Abdulla et al., 2001;

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Abdul Rahman et al., 2010b; Issa et al., 2011; Noor et al., 2010, 2011); electrons (Hashim et al., 2009; Abdul Rahman et al., 2010b); protons (Hashim et al., 2006); alpha particles (Ramli et al., 2009); neutrons (Hashim et al., 2010) and synchrotron radiation (Abdul Rahman et al., 2010a). The TL yield is dictated by the concentrations and types of intrinsic or extrinsic defects within the medium. In practice, variations in these are reflected in the sensitivity of the dosimeter (Chen and McKeever 1992; Youssef, 2003), a situation resolved by selection through sensitivity screening should the intention be to use the dosimeter in practice. Present studies represent the X-ray response of undoped flat fibres and commercial Ge-doped SiO₂ fibres, compared against the standard photon TL material, TLD-100, all unscreened.

Noting that Pt-based cisplatin drugs have immunochemo- and radio-therapeutic effects of use in improving loco-regional and systemic tumour control (Abundes-Velasco et al. 2007; Bhide et al., 2012; Guida et al., 2008), we further explore the potential of flat fibres for investigation of the high-Z photoelectron-generated local dose enhancement. In this we use nm-thick Pt-coatings to the silica fibre dosimeters. As an aside, the untoward effect observed in clinical use of X-rays of enhanced dose local to Pt implants (Cheung and Tang, 2007) is something that our fibres would also be able to measure.

Finally, we are seeking to explore the defects-basis of the SiO₂ TL underpinning our work. In this, we investigate the elemental, chemical and electronic state of the silica using X-ray photoelectron spectroscopy (XPS). This non-destructive surface-sensitive quantitative spectroscopic technique is based on the lossless emission of photoelectrons from the sample surface, the escape depth varying from 0.1 nm to 1 nm depending on the kinetic energy of the photoelectron. As such, the surfaces under study must be studied under stringently clean conditions.

2. Materials and methods

2.1. Samples and their preparation

We compare single-mode doped SiO₂ telecommunication fibres, undoped SiO₂ flat fibres and TLD-100 chips. For the first, use was made of INOCORP (Canada) fibres comprising a 9.0 µm Ge-doped core in a SiO₂ cladding of diameter (124.7 ± 1.0) µm plus a polymer coat of diameter (250 ± 10) µm. Following polymer removal the doped fibres have been cut into approximate 0.5 cm lengths. The undoped flat fibres have been fabricated using a 5 m fibre drawing tower located in the Department of Electrical Engineering, University of Malaya (Dambul et al., 2012). The flat fibres have been fabricated using a high grade amorphous SiO₂ tube (Suprasil F300), outer and inner dimensions 25 mm and 19 mm. As the SiO₂ tube preform is pulled down into capillary form (Fig. 1(left)) of around 1–2 mm diameter, vacuum pressure is applied to collapse the capillary into the flat form (Fig. 1(middle) and Fig. 2). The dimensions of the flat fibres, obtained by digital calipers (DURATOOL), are detailed in Table 1. The TLD-100 (LiF:Mg, Ti), comprises 99.99% ⁷Li, in the form of a disc of diameter 8.81 mm and 0.35 mm thickness. To avoid surface abrasion and

Fig. 1. From left to right are not-to-scale schematic representations of capillary fibre, flattened capillary fibre (flat fibre) and the common form of doped single mode telecommunication fibre (from Alawiah et al., 2013).

Fig. 2. Expanded schematic of pure silica flat optical fibre (from Alawiah et al., 2013).

Table 1

Mean dimensions of eleven undoped silica flat fibres (FF) (see Fig. 2).

Dimension	(mm)
a	1.13
b	1.00
W	3.72
L	10.00
Weight (g)	0.085

contamination, vacuum tweezers (Dymax 5, Surrey, UK) have been used to handle the TL materials. To minimize light exposure, all three types of dosimeter were kept in darkened conditions before and subsequent to irradiation, until readout.

2.2. Annealing

Before making irradiations, the samples (all unscreened) were first annealed in a furnace (Carbolite, UK). This was carried out to provide for a stable situation, for both background and TL signal, the annealing allowing elimination of previous irradiation history, standardizing the thermal history as well as sensitivity, also erasing the unstable low-temperature glow peaks. For annealing, the samples were retained in an alumina ceramic boat and covered with Al foil, annealing being performed at 400 °C for a period of 1 h. Subsequently, to minimize thermal stress, the samples were left in the oven for 18 h to finally equilibrate at room temperature.

2.3. Flat fibres coating

The undoped flat fibres were arranged to be Pt-coated on one side only (to allow read out using the uncoated side), use being made of a sputter coating unit (Emitech K575X) located at the Surrey Materials Institute. The fibres were coated with 20 nm, 40 nm, 60 nm and 80 nm of Pt. The sputter current was set to 70 mA and the 20 nm terminate value was selected. Upon each cycle completion, one of the fibres was taken out of the sputterer and the rest would be coated for another 20 nm. The procedure was repeated until all four thicknesses of coated fibres were obtained. Pressure sensitive adhesive (Blu-Tack) was used to secure the fibres on the specimen stage (glass slide).

2.4. Sample irradiation

For comparative sensitivity the samples were irradiated at the Royal Surrey County Hospital (RSCH), use being made of a ClinacTM 2100C linear accelerator (LINAC). The TL media was exposed to 6 MV X-rays to give a dose of 3 Gy. The source to sample surface distance (SSD) was set at 100 cm, with a field size of 20 cm × 20 cm. Subsequent to Pt coating the flat fibres were irradiated to tube X-rays produced at an accelerating potential of 250 kVp, again to a dose of 3 Gy. In all cases, the fibres were placed at the centre of the field, with the coated side facing towards the

X-ray tube, aligned with the applicator of the X-ray unit with a 3 cm stand off distance from the phantom to the applicator.

2.5. Readout

After irradiation, the samples were read out using a Toledo TLD reader (Vinten, Sandy, UK), with a N_2 gas atmosphere set at 0.5 bar to inhibit the effects of chemi- and triboluminescence. During readout, the following time-temperature cycle was used: a preheat temperature of 120 °C for 28 s; readout temperature of 300 °C for 25 s; a heating cycle rate of 38 °C/s. Lastly, an internal annealing temperature of 300 °C was used for 28 s to provide initial erasing of any residual signal. The arrangement provided for an optimal glow curve.

2.6. X-ray photoelectron spectroscopy (XPS)

This was carried out using an XPS Thermo Scientific Theta Probe located at the Structure Analysis Laboratory and Micro Structural Studies Unit of the University of Surrey. The sample analysis was performed in an ultra-high vacuum (UHV), the electron flux being evaluated with minimum error at each value of kinetic energy. The arrangement also minimises recontamination of the freshly cleaved sample, noting that in XPS instruments the electron counting detectors are typically 1 m distant from the sample. With a high voltage potential applied between the filament and anode to accelerate electrons towards the target, electron bombardment of the target produces core vacancies, causing photoelectrons and emission of fluorescence X-rays. The photoelectrons enter an energy analyzer coupled to an electron detector. The facility is typically operated at an anode voltage of 15 kV and a current of 20 mA to produce sufficiently intense photoelectron peaks. Prior to use, the sample holder, sample stub (ESCALAB) and fibres were cleaned using an ultrasonic bath, repeated using three different solvents; methanol, ethanol and propanol. To ensure minimal charge collection the stub was coated with ~ 50 nm gold.

3. Results and discussion

Using the 6 MV photon beam, eleven of each of the TLD-100 chips, Ge-doped fibres and flat fibres were simultaneously irradiated to obtain their comparative TL yield (Fig. 3). Interest focuses on the previously unexplored sensitivity of the flat fibres. Fig. 3 shows the much greater TL response of the flat fibres compared to the Ge-doped fibres, of the order of 100 times that produced by the Ge-doped fibres, being also similar to that from TLD-100. This is due in good part to the differential mass of the two dosimeter types, the Ge-doped fibre mass being approximately 1000 times

less than that of the flat fibre, and also due to the undoped nature of the flat fibres. In undoped optical fibres, the defect centres are mainly induced during the fibre pulling process (Friebele et al., 1976; Hibino and Hanafusa, 1988). Thus said, it is expected that pulling parameters such as vacuum pressure, pulling speed, high pulling temperature of above 2000 °C and fast cooling/quenching rate in room temperature play an important role in the TL response, influencing the species and population of defects and characteristics of optical fibre. The deformation of the circular shape into a flat shape produces stresses on the circumference of the neck-down region of the preform that generate additional structural defects. The shear stress applied to viscous state induces non-bridging oxygen hole centres (NBOHCs) and breakage of the strained Si–O bonds in the continuous random silica network defining the amorphous silica (Hibino and Hanafusa, 1988). The findings are supported by theoretical predictions (Roushdey, 2011).

The sensitivity of undoped flat fibres is increased through application of high atomic number coating to enhance photoelectron production. In this case, Pt ($Z=78$) was used as a coating material. The dose enhancement factor (DEF) is described as the ratio of dose deposited in the Pt-coated fibre to the dose deposited in the uncoated fibre. The results in Fig. 4 show a linear increase in dose enhancement factor (DEF) from 20 nm up to 80 nm. At 80 nm the DEF attains its highest value of 1.20 ± 0.03 , leading to a summary DEF of $(0.0150 \pm 0.0003) \text{ nm}^{-1}$. The result is comparable to the previous study of Alalawi et al. (2013) using gold, which showed a DEF of $(0.0160 \pm 0.0001) \text{ nm}^{-1}$. The difference of 6% between the DEF of Au and Pt coatings is in line with the photoelectric effect probability difference of 5%.

In XPS, atomic concentrations (with a lower limit of approximately 0.1 at%) can be quantified. Fig. 5 shows the presence of a number of elements on the sample surface, the different peaks corresponding to electrons photoejected from the different core levels of the material. A higher resolution scan is observed in Fig. 6, showing the Si2p core level peak. In essence, the peak positions reflect binding energies, providing information about the chemical state for a material. The peak binding energy of Si2p is at 103.86 eV. The atomic concentration of the Si2p state obtained from the XPS analysis is found to be 22.41%.

4. Conclusion

In present study, the sensitivity of the undoped flat fibre has been found to be markedly higher (by a factor of 100) than that of Ge-doped fibre, being comparable to that of TLD-100. The study results are suggestive of defects generation occurring as a result of the collapsing technique in producing the flat fibres, providing a TL response from the optical fibres that can improve upon existing TL system sensitivities. Following on from this, investigation has been

Fig. 3. Logarithmic plot of the TL yield distribution for flat fibres, TLD-100 chips and Ge-doped fibres subjected to 6 MV photon irradiation. (Note: the error bars are smaller than the data points).

Fig. 4. Dose enhancement factor (DEF) for different thicknesses of platinum coating.

Fig. 5. XPS survey spectrum of an undoped flat fibre, analysed over the cross-section of the fibre using a 400 μm focal spot.

Fig. 6. High resolution scan of Si2p core level peak.

made of the effect of different thicknesses of platinum coating on the thermoluminescence response of the tailor-made undoped SiO_2 flat fibres. The results show a dose enhancement factor of $(0.0150 \pm 0.0003) \text{ nm}^{-1}$, which is comparable to that obtained using gold-coated fibres, the latter validated by Monte Carlo simulations. Information of the identification of elements near the surface and oxidation states of transition metals has been investigated using the XPS method. The charge state of Si2p was found to be associated with a binding energy of 103.86 eV, the atomic percentage of the Si2p state being 22.41%.

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