Radiation dosimetry using optically stimulated luminescence in fluoride phosphate optical fibres

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Abstract: Optically stimulated luminescence has been demonstrated within a fluoride phosphate glass optical fibre. These fibres have been used as the basis of a novel dosimeter architecture whereby the optical fibre acts as both the sensing and light guiding component. Fibres were fabricated from a commercially acquired glass and irradiated using a $^{90}Sr/^{90}Y$ source. Following optical stimulation with a 532 nm laser, optically stimulated luminescence of 483 $\pm$ 18 cnts/g/μJ was detected. In addition to OSL, scintillation of 6155 $\pm$ 78 cnts/s was also measured. A linear response between 0.16 - 2 Gy of irradiation was measured, after which the intensity was observed to plateau. These results indicate the potential suitability of fluoride phosphate optical fibres for radiation sensing applications.

OCIS codes: (060.2290) Fiber materials; (060.2370) Fiber optics sensors; (160.2540) Fluorescent and luminescent materials; (160.2750) Glass and other amorphous materials; (260.3800) Luminescence.

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1. Introduction

Optical fibre dosimetry has been studied as a method of monitoring radiation for medical in-vivo applications and distributed measurements in environments of high dose-rate [1, 2]. Optical fibres provide several advantages in the field of dosimetry. They are lightweight and non-intrusive, which is beneficial for medical applications. The response is optical, and therefore results are obtained immediately without the need for analysis which, for some devices such as thermoluminescent dosimeters, can take several hours. They are also capable of remote sensing, and are suitable for use in confined environments that may be inaccessible using existing dosimeters.

Optical fibres can be utilised for radiation sensing using two methods. The first employs an extrinsic architecture, where the radiation-sensing component is spliced or coupled to an optical fibre [1, 2]. In such cases the fibre acts only as a waveguiding component to carry an optical signal from the sensing component to a detector. Scintillation and optically stimulated luminescence are commonly utilised for this sensing architecture. Two examples of materials used as the radiation-sensing component are Cu^+-doped silica [3] and $Al_2O_3 : C$ [4]. Recent work with these materials demonstrates the ability of this sensor architecture to perform non-intrusive, in-vivo monitoring during radiotherapy [5, 6].

The second type of fibre radiation sensor has an intrinsic architecture, where the optical fibre is used as the radiation-sensitive material, in addition to guiding the optical signal to the detector. This architecture is more suitable for environmental monitoring, or applications requiring monitoring over a long distance, rather than a single point. To date, this architecture has been demonstrated based on the mechanisms of scintillation and photodarkening. Scintillation was first studied in optical fibres by Marrone et. al. in 1981 [7], photodarkening was first studied in fibres by Evans et. al. in 1978 [8]. An example of recent research utilising scintillating fibres for monitoring purposes can be found in [9], in which polymer optical fibres were positioned around containers of radioactive waste in order to monitor gamma emission. An example of recent developments into the photodarkening technique involves the radiation-induced attenuation of polymer fibres when exposed to gamma radiation between 12-34 Gy [10]. This work has shown that photodarkening at one wavelength can be used to continuously measure the absorbed dose of the material in real-time.

In this study we focus on advancing the intrinsic fibre architecture, for the purposes of distributed and environmental sensing in situations where an increase in the radiation field is possible. There are many potential environments that would benefit from this method of sensing, primar-

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ily any environment in which leakage of ionising radiation from equipment or facilities could potentially occur [11].

The primary sensing mechanism used here is optically stimulated luminescence (OSL), and this is the first work that explores OSL in an intrinsic fibre sensing architecture. We also measure the scintillation response of the fibres and consider how it may be used to support OSL measurements. As mentioned briefly, OSL is used in chip-coupled optical fibre sensors, but has not previously been used in a distributed fibre sensing device. Huston, et. al. (2002) described a glass capable of producing optically stimulated luminescence [12], however it was not used in an optical fibre configuration, but instead was spliced to the end of non-active commercial fibre for the purposes of point measurements. This paper builds upon previous work whereby fluoride phosphate glasses were developed as strongly OSL-active materials [13]. Here we briefly describe the fabrication of this material into optical fibres. Following this, we present the measurements of the radiation sensing properties of the fibres. First, a brief introduction to the theory of optically stimulated luminescence is provided.

2. Optically stimulated luminescence

Optically stimulated luminescence [14, 15] occurs when ionising radiation interacts with a bandgap material, such as glass. Energy is imparted to electrons, exciting them into the conduction band. If the material has suitable traps, these excited charges may be trapped in a metastable state on the scale of seconds to millions of years, depending on the depth of the trap. To induce the stimulated luminescence and 'interrogate' the material, it is exposed to light with sufficient photon energy to excite electrons out of the traps. Untrapped electrons are therefore able to move through the conduction band from where they can be captured at recombination centers, this relaxation may occur via a radiative transition thus producing detectable luminescence. In a glass material, traps are typically formed by anion vacancies in the lattice, such as an anion or fluoride ion vacancy. Radiative recombination will generally occur in a metal ion impurity, such as a dopant ion, and as such the wavelength of luminescence may be tailored to a specific region. Figure 1 shows each step in the process.

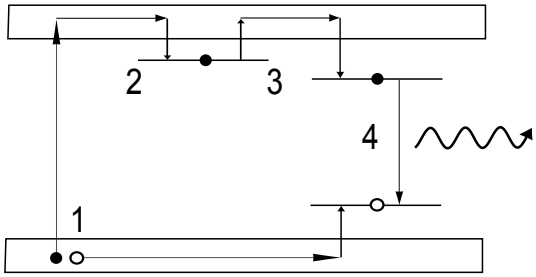

Fig. 1. Fundamental mechanism of optically stimulated luminescence: (1) Ionising radiation creates an electron-hole pair in the glass (2) electron is trapped in an excited state (3) optical stimulation releases this trapped electron (4) electron is free to recombine with a hole via a radiative transition.

3. Material selection and fibre fabrication

Fluoride phosphate glass was previously identified as a suitable material for sensing ionising radiation by the mechanism of optically stimulated luminescence [13]. A commercially manufactured fluoride phosphate glass, Schott N-FK51A [16], was also identified as having a similar

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glass composition. This commercial glass has a similar OSL response with glasses produced in-house, and was therefore used for fibre fabrication.

Optical fibre preforms were prepared using the extrusion method [17]. Bare, unclad fibre preforms (i.e. rods) of 10 mm diameter and approximately 200 mm in length were produced by extrusion through a Macor® ceramic die at 525 ºC. This temperature range and die material produced a good preform surface quality when compared to other die materials, such as stainless steel and graphite. Fibres were drawn directly from extruded preforms on a drawing tower, using a furnace temperature of 753 ºC (approximately 550-600 ºC glass temperature) and a preform feed-rate of 4.5 mm/min. Unclad bare fibres of 160 μm diameter were produced. The transmission loss of the fibre was measured using the cutback method. Loss measurements were performed at 405 nm, the wavelength of interest for luminescence measurements. Fibre quality was found to vary along the fibre length, leading to uncertainty in the transmission loss measurements. A value of approximately 10 dB/m was obtained, however transmission quality was measured to vary between 4-14 dB/m. The transmission of bulk glass at 405 nm has been quoted at 1.22 dB/m by the manufacturer [16], future work will focus on achieving similar transmission quality in the optical fibres.

4. Optically stimulated luminescence measurements

The experimental setup used to measure the dosimetry properties of the fibres is shown in Fig. 2. By using stacked fibre bundles the cross-section of material exposed to radiation, and therefore the OSL signal, can be increased. Therefore a bundle of 29 individual fibres, each 1.2 m in length, was arranged into a coiled bundle of 45 mm radius, which passed beneath the radiation source three times. Based on these dimensions, the irradiated mass of material was 64.4 mg. A ^{90}Sr/^{90}Y beta source was used for irradiation, delivering a nominally calculated dose rate of 2 Gy/min. Calculation of the exact dose-rate is being performed using Geant4 software and will be reported in future results. A 532 nm laser was used for stimulation of luminescence. The delay between shutting the radiation source and applying laser stimulation was kept constant at three seconds. The optical stimulation power was calculated to be 78.5 μW within the 64.4 mg of radiation-exposed mass. Detection of luminescence was performed using an EMI 9635 QA photomultiplier tube. Isolation of the luminescence from the stimulation light was performed using a dichroic mirror at 505 nm and two 4 mm Corning 7-59 filters. The bandpass filters were chosen based on the quantum efficiency of the photomultiplier tube in the 300 - 550 nm region. The laser was selected at a wavelength short enough to provide effective stimulation of trapped electrons, but a wavelenth long enough such that it was sufficiently attenuated by the filters. The selection of wavelengths is illustrated in Fig. 3, which shows the transmission of the Corning 7-59 filters, quantum efficiency of the photomultiplier tube and the laser stimulation wavelength.

An example of the measurement cycle can be seen in Fig. 4, where a dose of 4 Gy was applied to the fibre bundle. Data acquisition was started prior to removing irradiation and therefore the scintillation could be measured. Scintillation was quantified by integrating over the first shaded region shown in Fig. 4, this was normalised to the mass of the irradiated section of the fibre bundle, 64.4 mg. In the measurement shown, scintillation was measured to be 95574 $\pm$ 1211 cnts/s/g. The detected scintillation potentially offers information on the strength of the ionising radiation and dose-rate, however it does not yet imply any energy resolution or identification of the type of ionsing radiation.

After irradiation, the radiation source was switched off so as to remove any scintillation from the background when measuring the OSL signal. Laser stimulation was then applied and the OSL measured. In the example shown in Fig. 4 the intensity is measured at 85 counts, after which it decays to the background as the population of trapped electrons is depleted. This re-

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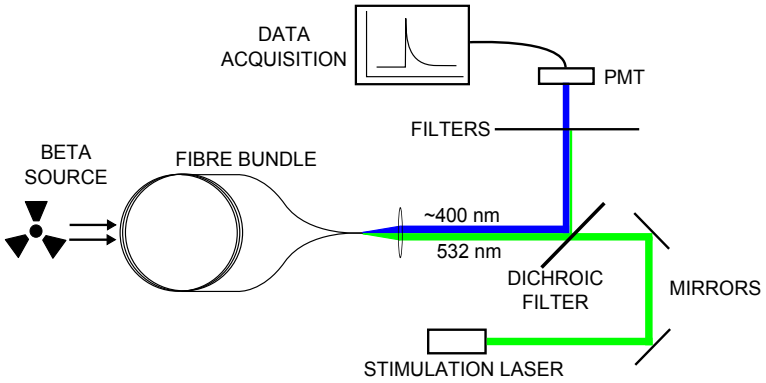

Fig. 2. Experimental configuration used for the detection of optically stimulated luminescence in optical fibre bundles. Optical stimulation is at 532 nm, luminescence is produced at approximately 400 nm.

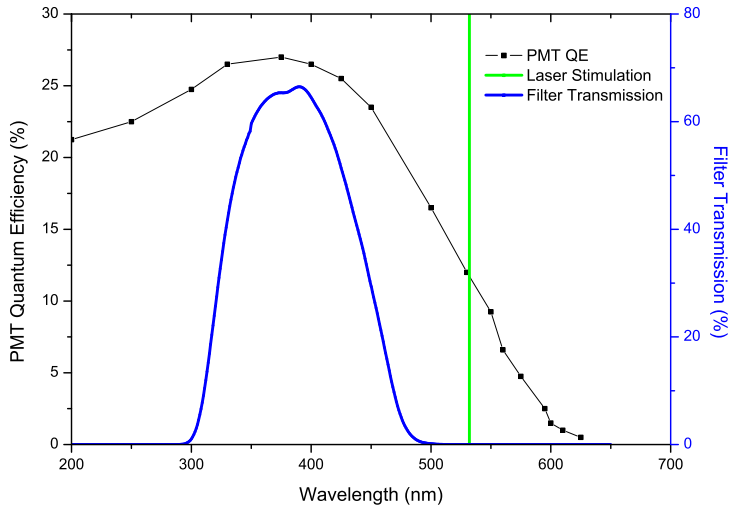

Fig. 3. Stimulation and detection wavelengths used for OSL measurements. Shown are the quantum efficiency of the EMI 9635 QA photomultiplier tube, the transmission of the Corning 7-59 band pass filters, and the laser stimulation wavelength of 532nm.

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sponse is typical of OSL [14,15] and indicates the presence of an OSL response from the optical fibres. The OSL is quantified by integrating from the initial laser stimulation until the majority of the signal is removed, shown by the shaded region in Fig. 4 at 11.5 seconds. The background is also measured by integrating the counts once the OSL signal has been removed and subtracted from the luminescence response, again shaded in Fig. 4. This background is comprised of the PMT dark count, a small anti-stokes component, and a slight contribution at 532nm from laser light which penetrated the filters. Values are normalised for the laser stimulation energy of 15.7 μJ and the mass of the irradiated section of the fibre bundle, 64.4 mg. Measurements were taken several times and the results averaged, for the measurement described here the average value obtained was an intensity of 483 $\pm$ 18 cnts/g/μJ.

The quantification of both scintillation and OSL within the one measurement cycle offers interesting possibilities for the monitoring of ionising radiation. One method might be to continuously monitor a scintillation signal to obtain the dose-rate, while periodically interrogating the fibre for an OSL response, which would yield information on the dose absorbed over a certain time period. The combined use of both these luminescence mechanisms will offer more information than devices using only one or the other.

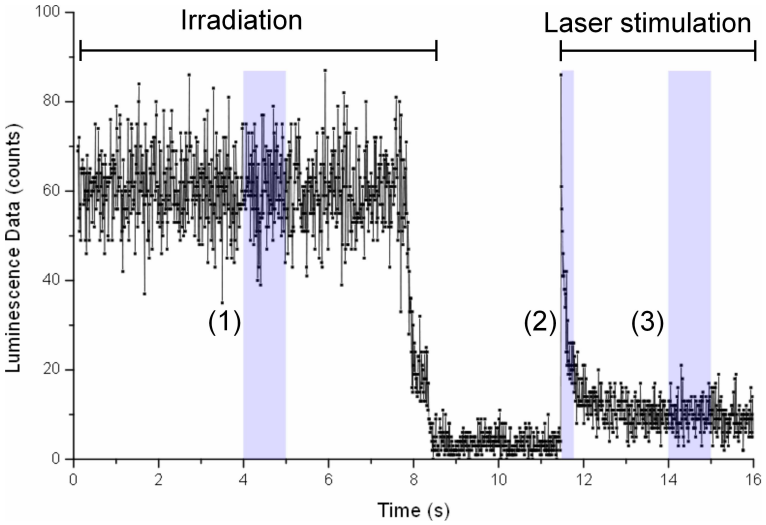

Fig. 4. Luminescence with respect to time during the measurement of OSL. Shaded regions indicate where integration of photon counts was performed in order to quantify (1) scintillation, (2) OSL and (3) the background. Channels are of 10 ms duration.

The OSL measurement described was repeated over a short time period to determine the cumulative effects of re-exposing the fibres. Samples were irradiated with an identical dosage of 1 Gy after which the OSL was measured. Following the OSL signal, a short period of laser stimulation was found to completely remove any charge remaining in a metastable state, thus 'bleaching' the material back to the state it was in prior to irradiation. This was performed for ten consecutive measurements and can be seen in Fig. 5. Following repeated dose/readout cycles, the intensity of the OSL response was found to be constant at 219 $\pm$ 10 counts/g/μJ.

This result indicates efficient bleaching of the material. If the laser stimulation is left on for a short period of several seconds following the OSL measurement, any excess charge still in an excited state will be removed. Therefore the material is 'reset' before the next measurement and no detectable residual dosage information remains to affect the next measurement. This is an important property of the fluoride phosphate glass when compared to other materials used

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for OSL, such as $Al_2O_3:C$, which require significant optical bleaching before re-use [18]. This efficient bleaching of excited charge may be attributed to shallow traps, from which electrons can be stimulated quickly. The thermoluminescence produced by this material exhibits only short-lived glow peaks, with lifetimes of several minutes, consistent with only shallow traps being present.

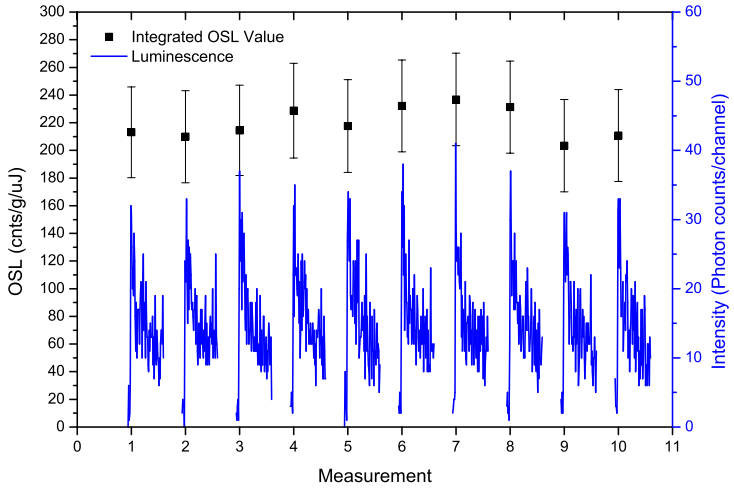

Fig. 5. Reproducibility of OSL response over ten consecutive measurements. Original luminescence data is shown in conjunction with the integrated value, where the time interval shown for each measurement for each is 0.65 s.

The OSL response of the fibres with respect to applied dosage was also measured. Using the same experimental conditions as above, the dosage applied to the bundles was increased from 0.16 - 16 Gy. The activity of the $^{90}Sr/^{90}Y$ beta source is constant, so the total dosage was varied by increasing the time over which the fibres were exposed. Figure 6 shows the relationship between applied dosage and the measured OSL. A linear relationship is observed between 0.016 and 2 Gy, after which the intensity no longer follows this trend. Previous measurements on this glass show saturation of the material at approximately 2 Gy [13], this saturation was attributed to complete filling of available traps, such that there was a limit on the population of excited charge. The saturation of the OSL signal observed from fibres may be attributed to this effect, but it may also be attributed to the short life-time of trapped charge. The time required to expose the fibres to a dosage of 2 Gy is 60 seconds, which is similar to the lifetime of the trapped charge, such that a dynamic equilibrium is established. Above 8 Gy the OSL response is seen to decrease, which is consistent with the behavior observed in other luminescent materials such as $Al_2O_3:C$ [19]. This behavior has been studied theoretically and has been attributed to two possible mechanisms [20]. The first involves electrons competing for certain traps during irradiation at high dose rates, of which only a fraction of the traps satisfy the conditions for optical stimulation. The second involves competition for recombination centres during optical stimulation of a large population of excited charge, where only a fraction is able to recombine through a radiative transition.

Experiments were performed to determine the minimum number of fibres necessary for OSL detection. For this example, a radiation dose of 8 Gy was used based on the results illustrated in Fig. 6. The experiment involved taking a number of OSL measurements, where each successive measurement was performed with one less fibre in the bundle, results are shown in Fig. 7. The

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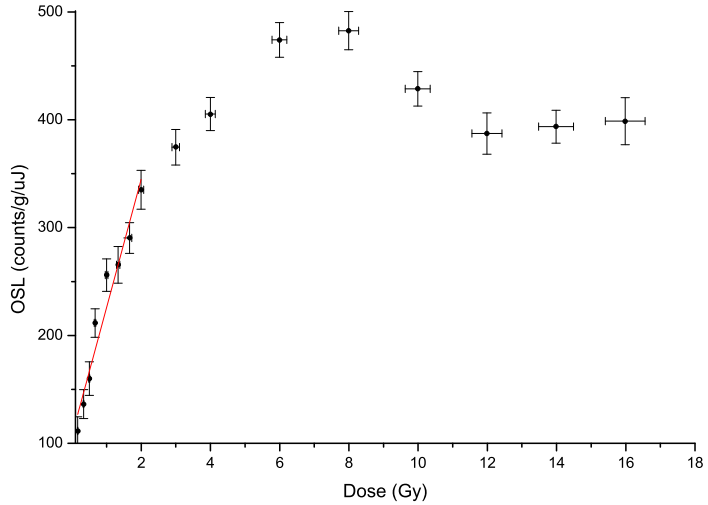

Fig. 6. OSL intensity with respect to the applied radiation dose. A linear trend is observed between 0.016 and 2 Gy, with an R^2 value of 0.934.

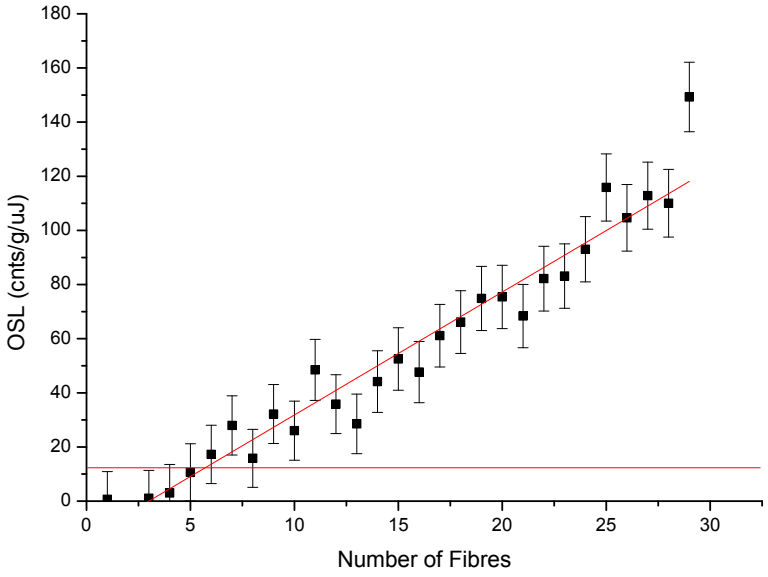

Fig. 7. OSL response with respect to the number of fibres exposed to 8 Gy of radiation. Current fibre quality requires approximately six fibres to observe the OSL signal over the noise at this radiation dosage.

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limit of OSL detection was defined as the average uncertainty for an OSL measurement, 11.3 cnts/g/μJ, and is shown in Fig. 7 by the horizontal line. The number of fibres required for OSL detection was defined as the intercept between this line and the linear fit of the OSL response. For 8 Gy of radiation, this was determined to be six, as can be seen in Fig. 7, which is equivalent to an effective irradiated mass of 13.3 mg. Predictions indicate that if the loss is reduced to 1.22dB/m, the transmission of the material at 405nm, detection of OSL will be possible with 4 fibres. Furthermore if the background noise, and hence the uncertainty, is reduced, detection with 1-2 individual fibres is theoretically possible.

5. Conclusion

For the first time, optical fibres were used simultaneously as a radiation detecting medium using optically stimulated luminescence, and as the signal-guiding medium. An OSL response of 483 $\pm$ 18 cnts/g/μJ has been measured. Additionally, scintillation has been measured at 6145 $\pm$ 78 cnts/s in the same fibres.

A linear relationship between the applied dosage and the intensity of the detected OSL signal has been measured between 0.16 and 2 Gy. Efficient optical bleaching of the material has been observed, such that measurements can be repeated immediately without any residual signal or cumulative effects being observed. Detection of 8 Gy of radiation was possible with a bundle of six fibres, whereas an intensity of 17 cnts/g/μJ was measured. Improvements in fibre quality, material composition and detector sensitivity could potentially increase the sensitivity such that a single fibre could be used for detection. The results presented here demonstrate the potential of fluoride phosphate optical fibres for environmental monitoring in situations where elevated dose-rates may occur.

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