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ARTICLE

Ion Beam, SEM and EDXRS Analysis on Doped SiO_2 Optical Fibres

Suhairul HASHIM[1*], David Andrew BRADLEY[2], Ahmad Termizi RAMLI[1], Husin WAGIRAN[1], Melanie WEBB[3], Chris JEYNES[3], Mohd. Iqbal SARIPAN[4], Mohd. Nadzri MD REBA[5] and Khalid ALZIMAMI[6].

[1]*Department of Physics, Universiti Teknologi Malaysia, 81310 Skudai, Johor, Malaysia.*
[2]*Department of Physics, University of Surrey, Guildford, GU2 7XH, U.K.*
[3]*Surrey Ion Beam Centre, University of Surrey, Guildford, GU2 7XH, U.K.*
[4]*Faculty of Engineering, Universiti Putra Malaysia, 43400 Serdang, Selangor, Malaysia.*
[5]*Faculty of Geoinformation and Real Estate, Universiti Teknologi Malaysia, 81310 Skudai, Johor, Malaysia.*
[6]*Applied Medical Sciences College, Salaman Bin Abdulaziz University, PO.Box 422, Alkharj 11942, Saudi Arabia.*

Optical fibres have been demonstrated by this group to show promising thermoluminescence (TL) properties with respect to ionizing radiation for low-energy X-rays, megavoltage photons, -rays, accelerated electrons and accelerated protons, α-particles and fast neutrons. Present research has focused on commercially produced single-mode telecommunication optical fibres manufactured by INOCORP (Canada). These being either in the form of SiO_2 optical fibres doped with Ge or Al at concentrations appropriate for total internal reflection, as required for telecommunication fibres. It is to be noted that dopant concentrations are not included among the data provided in the accompanying product data sheets. Each of these INOCORP fibres has a core diameter of 125.0 ± 0.1 μm. To achieve this aim using ion beam analysis, PIXE (Proton Induced X-ray Emission) which, while having limited depth resolution, can unambiguously identify elements and analyse for trace elements with detection limits approaching μg/g. For Al-doped fibres, dopant concentration in the range 0.98 - 2.93 mol % have been estimated, the equivalent range for Ge-doped fibres being 0.53 - 0.71 mol %. Based on SEM (Scanning Electron Microscope) and EDXRS (Energy Dispersive X-ray Spectroscopy) analysis, the Al-doped fibres were evaluated to have dopant concentrations in the range 0.35 – 0.88 mol %, the equivalent range for Ge-doped fibres being 0.15 - 0.19 mol %.

KEYWORDS: Thermoluminescence(TL); SiO_2 optical fibres; dopants; PIXE, SEM and EDXRS analysis

I. Introduction

It is extremely useful to have a sensitive, robust and high spatial resolution system for radiation measurement to be used in medical application. Ion beams are used in radiotherapy to deliver a more precise dose to the target volume while minimizing a dose to the surrounding healthy tissue. For optimum dose monitoring in ion beam therapy, it is essential to be able to measure the delivered dose with a sensitivity, spatial resolution and dynamic range that is sufficient to meet the demands of the various therapy situations. Thus said, in terms of current radiotherapy practice relevant to dose monitoring, the use of ion beams remains relatively small.

Compared with the use of TLD phosphors such as LiF:Mg,Ti and LiF:Mg,Cu,P, the fibres not only offer the possibility of improved positional sensitivity (fibre diameters are sub-millimeter, typically ~ 200 μm) but also, since the fibres are impervious to water (the silicates forming a glass in the fibre-preforming process), this paves the way for their use in intercavitary and interstitial measurements[1]. In patients with severe medical problems for whom few, or no, alternative diagnostic techniques can be envisaged, doses have been delivered to an extent resulting in severe skin necroses[2]. Geise and O'Dea[3], cite doses of several tens of Gy to the skin in several such medical investigations, also reviewing moves towards ensuring skin dose reduction in so far as this may be possible for the particular situation. At this point, it is sufficient to point out that there are many interesting dosimetric applications of optical fibres that could be imagined in such situations.

Optical fibres have been demonstrated by this group to show promising thermoluminescence properties with respect to ionizing radiation for photon[4,5], electron[4,5] alpha particle[6] and fast neutrons[7] irradiation.

The interest of our group is to analyze and verify the performance of commercially produced doped SiO_2 optical fibres with a view to improving the thermoluminescence (TL) yield. The measurements have been conducted after removing the plastic coating of the optical fibre to allow elemental analysis to be done using PIXE and RBS techniques on the core of the fibres. It is intended that these efforts will lead to the design of *in situ* doped silica fibre dosimeters for particular applications in radiotherapy, high-dose radiodiagnosis and industrial/nuclear industry settings.

II. Materials and methods

1. Sample preparation

In preparation for irradiation, the outer polymer coating to

*Corresponding Author, E-mail:suhairul@utm.my

the optical fibres was removed using a fibre stripper (Miller, USA) to allow investigation of the TL yield of the fibre core. Following removal of the outer cladding, the optical fibre was cleaned by means of a cotton cloth containing a small amount of methyl alcohol to completely remove any remnant polymer cladding. Subsequently, the fibre was cut into 0.5 cm long pieces using an optical fibre cleaver (Fujikura, Japan). The mass of each fibre, ~ 0.20 ± 0.02 mg, was measured using an electronic balance (PAG, Switzerland). Vacuum tweezers (Dymax 5, Surrey, UK) was used for handling and grouping of the TL materials.

2. Ion beam based elemental mapping

The University of Surrey hosts the EPSRC (Engineering and Physical Sciences Research Council) national ion beam facility. The microbeam facility is based on a 2 MV Tandetron™ accelerator. A review of the facilities offered by the Surrey ion beam centre is covered in detail by Simon *et al.*[8].

In the present investigation, protons with energy of 2.5 MeV were used. For the PIXE and RBS analysis conducted, the scanning area was chosen to be 0.5 mm x 0.5 mm with a spot size of 3 μm x 5 μm and the beam current was ~ 70 pA. The scanning was done over the freshly cleaved cross-sectional area of the optical fibre samples, producing a pixel by pixel map of the sample elemental composition.

The copper alignment block acts as a drain for beam current and further allows the use of Cu Rutherford Backscattering Spectrometry as a reference against which O_2 data can be compared. During proton irradiations for PIXE and RBS analysis, the optical fibres were sandwiched in a parallel orientation and held between two customs made copper blocks.

Fig. 1 Magnified image of fibres on beam line as viewed using a microscope and monitor display.

In **Fig. 1**, sample positioning is carried out using a video microscope in combination with an alignment laser to ensure that the sample is reproducibly positioned at the correct distance from the exit window. The sample is illuminated using a fibre optic illuminator. At present, the sample is mounted on a manual micrometer positioning stage with a total range of movement of 20 cm on each axis. The pointed markers were used for easy viewing and to make sure the irradiation was performed on the correct fibres.

The ion beam facility has two beam lines: a microbeam (in vacuum) line and a millibeam (in air) line, the latter allowing study of wet samples. At each line, PIXE can be used to analyze a wide range of trace elements. The great advantage of the PIXE method is the possibility of simultaneous multi-elemental analysis, a short time of data collection and relative simplicity on sample preparation. In many cases PIXE analysis may be considered to be non-destructive.

Characteristic X-rays produced during the process of PIXE analysis provide information on the relative distribution of Ge and Si concentrations. For O_2, use was made of RBS spectroscopy when ion beam is directed to the sample, enters the sample, scatters on atomic nuclei and travels back out to be detected by a silicon surface barrier detector to show an energy distribution. RBS also enables us to answer the fundamental quantitative characterization questions i.e. what is in the sample, how much is there and how deep is it in the sample.

It is important to note here that the dopant distribution in the core and inner cladding of a single-mode optical fibre determines the optical transmission properties of the fibre. If the fibres are exposed to high temperature for a period of time during their production, then the dopant distribution in these regions can change due to dopant diffusion. During the optical fibres fabrication process, the dopants i.e. Ge was added with a certain amount. The dopant tends to diffuse in the core and inner cladding of an optical fibre will determine the transmission properties of the fibre. Germanium is the dopant that has been most frequently reported in diffusion studies during optical fibres production. Diffusion of Ge in silica optical fibre also has been observed during splicing, manufacture of fused fibre couplers and fibre drawing[9].

3. SEM and EDXRS analysis

This was performed in the Faculty of Mechanical Engineering, Universiti Teknologi Malaysia, the particular Faculty hosting a Scanning Electron Microscope (SEM) facility based on GEMINI technology (Zeiss, Germany). The SEM technique was used to determine the effective atomic number of the Ge-doped optical fibres by measuring the relative elemental composition. The optical fibres were attached to the surface area of a stainless steel sample holder, electrostatic effects having first been dealt with by gold coating the fibre samples by a sputtering procedure. The scanning process was performed over the surface and the cross-sectional area of the fibres. INCAEnergy software (Oxford Instruments, U.K) was used to identify the elements present in the optical fibres. The X-ray spectrum was obtained from the X-rays being emitted from the samples. The software automatically identifies the peaks in the spectrum and the elements in the sample. Elemental presence can also be detected manually if the optical fibres' composition was known. The quantity of the element is calculated automatically.

4. Effective atomic number, Z_{eff}

Control of radiation dose is essential in performing an experiment in a biomedical context. One important factor in this is the tissue equivalence of the dosimetric material. Mixtures or compounds that are similar in their radiation

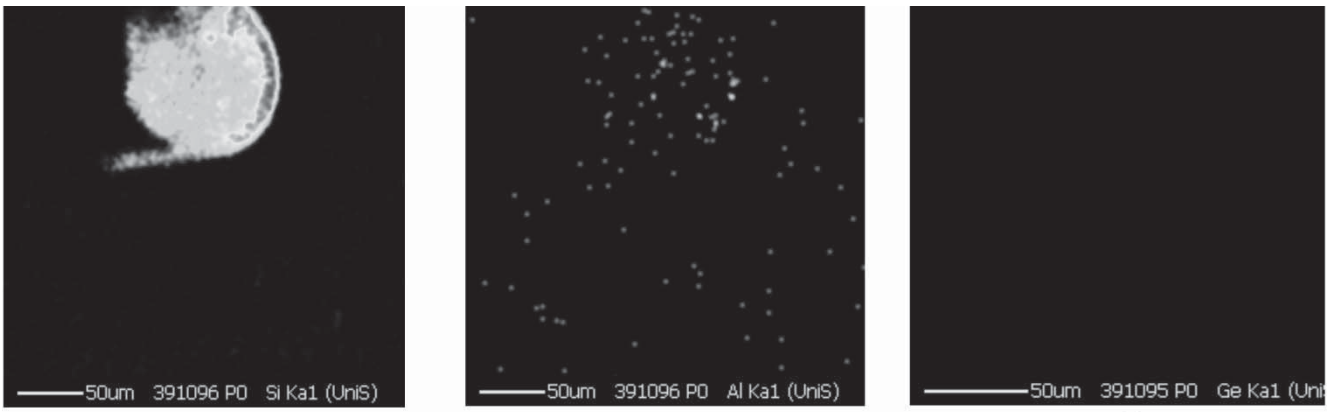

Fig. 2 (a) 300 μm maps of the cross section of the SiO_2 optical fibre (b) The presence of Al dopant (c) No Ge is seen on the map.

interaction characteristics to the soft tissue, bone or any other body constituents can be identified for this purpose.

The effective atomic number, Z_{eff}, of any mixture can be defined in terms of a single index for a given composite material, the Mayneord equation being used for this, with the estimate of Z_{eff} being given by:

$$Z_{\text{eff}} = (a_1 Z_1^m + a_2 Z_2^m + a_3 Z_3^m + ... + a_n Z_n^m)^{1/m} \quad (1)$$

where $a_1, a_2, a_3, ... a_n$ are the weight fraction contributions of each element in the fibre to the total number of electrons in the mixture.

Effective atomic number of a medium prescribes its detection efficiency and tissue equivalence. To obtain the effective atomic number of the doped fibres, SEM and EDXRS analysis was performed. The composition of the element inside the optical fibres was determined by SEM and EDXRS analysis based on the information given by the manufacturer (Oxford Instruments NanoAnalysis). A sputtering machine was used for the gold coating procedure, the gold being deposited in order to allow leakage of charge build-up to minimize the electrostatic effect during the scanning process.

III. Results and discussion

1. Determination of dopant concentration using PIXE and RBS analysis

The main advantages of PIXE and RBS analysis lies in its high sensitivity, possibility of simultaneous multi-elemental analysis of element of atomic number $Z \geq 11$ at all concentrations, its non destructive nature, the fact that no sample preparation is needed, the short time required for data collection (typically from a few seconds up to a few minutes) and the small sample area required, allowing good elemental concentration details to be examined.

In present studies as shown in **Fig. 2**, for Al-doped fibres, dopant concentrations in the range 0.98 – 2.93 mol % have been estimated, the equivalent range for Ge-doped fibres being 0.53 - 0.71 mol %. PIXE and RBS analysis also detected the presence of other elements, including P, Fe, Cl and Cu. However, the concentration of these elements was relatively small and can be considered to have negligible effect upon the TL yield of these fibres.

The main uncertainties in the absolute quantitation of PIXE elemental concentrations are the total deposited beam charge and the local matrix composition. The use of simultaneous RBS analysis can overcome this problem by providing the ratio between the true charge and the measured charge (the 'q factor'). After obtaining the 'q factor', the PIXE data can be normalized to provide accuracy of 5-10% in most cases. Details of the acquisition system have been described by Grime and Dawson[10] and Grime[11].

Results from these investigations were compared against previous studies by members of this group for samples of doped silica glass (with dopant concentrations from 0.01 mol % up to 33 mol %), produced using the sol-gel route. It was found from the previous study by Yusoff *et al.*[12] that the highest TL yield for Al and Ge doped fibres was at 4.0 mol % and 0.25 mol % respectively.

2. Determination of dopant concentration using SEM and EDXRS analysis

In present investigations of commercially available optical fibres, based on SEM and EDXRS analysis, the Al-doped fibres were evaluated to have dopant concentrations in the range 0.35 – 0.88 mol %, the equivalent range for Ge-doped fibres being 0.15 - 0.19 mol %. It would appear therefore that Ge-doped commercially available optical communication fibres offer close to optimal TL sensitivity while that of Al is sub optimal based on the previous results obtained[12].

3. Effective atomic number, Zeff

The value of Z_{eff} were determined using SEM and EDXRF analysis to be in the range of 11.5 - 12.1 and 11.3 – 13.7 for Ge-doped and Al-doped fibres respectively (value of Z_{eff} in soft tissue is 7.5). The value of m adopted in equation 1 for photon practical purposes is 2.94. These results indicate that Ge- and Al-doped are not tissue equivalent and as such the assessment of dose deposition in tissue would need to be corrected for this during the calibration process.

IV. Conclusion

Use has been made of PIXE, RBS, SEM and EDXRS analysis to map the relative presence of Al, Ge, Si and O. It is known that the pure silica will give rise to a degree of thermoluminescence (TL) following irradiation by ionizing radiation, the TL signal is considerably enhanced by the presence of certain dopants. The exact amount of dopant added to these fibres is not specified by the optical fibres manufacturers. The dopant acts as the defect centres that provide the TL signal. A particular concern is that non-uniformity in the distribution of added dopants and impurity concentration in the core of the commercially available fibres will cause variation in TL yield to an extent that this may limit the application of such fibres. Findings from these studies may pave the way to conduct more comprehensive investigation of TL from tailor-made doped SiO_2 optical fibres.

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