POLYMER OPTICAL FIBRE FOR IN SITU MONITORING OF GAMMA RADIATION PROCESSES

S. O'Keeffe, E. Lewis

Optical Fibre Sensors Research Centre, University of Limerick, Limerick, Ireland

Emails: sinead.okeeffe@ul.ie

Abstract- Poly Methyl MethAcrylate (PMMA) based plastic optical fibres offer a low cost radiation monitoring solution with a wide dose range, capable of providing real-time, online information. The sensitivity of the fibre to ionising radiation is shown to be dependant on wavelength, with the sensitivity increasing with decreasing wavelength, and so by carefully selecting the monitoring wavelength, the desired sensitivity and dose range can be achieved. The fibres exhibit high sensitivity, up to 0.6dBm^{-1}/kGy, and are capable of monitoring dose ranges between 30Gy and 45kGy. This exceeds the sensing range of all commercially available PMMA slab sensors, allowing it to be used for a wide range of applications.

Index terms: Radiation dosimetry, Plastic Optical Fibres, Optical Fibre Sensors, radiation processes

I. INTRODUCTION

Optical fibres provide the means whereby real-time in situ monitoring of radiation doses can be realised. They offer numerous advantages over conventional methods, such as electrochemical and semiconductor sensors, for sensing technology. Of significant advantage is the optical fibre's immunity to electromagnetic and chemical interferences and their ability to monitor remotely, whereby the sensor can be placed several hundred metres from the control electronics. This means that they can be employed in harsh environments, such as in high-radiation-level areas in the vicinity of a nuclear reactor or gamma sterilisation facility. PMMA based plastic optical fibres have a number of additional advantages, which make them particularly suitable for sensor systems, and with considerable improvement in transmission losses in recent years, research in this area has intensified [1]. Due to the large fibre cross-section of plastic optical fibres, generally a 1mm core, connecting to the light source and detector is non-problematic. This means that, in contrast to glass optical fibres, no expensive precision components are required for centring the fibres. The large fibre core diameter also means that more light can be transmitted and subsequently incident on the detector, resulting in a higher sensitivity of the sensor. Minor contamination, e.g. dust on the fibre end face, does not result in the complete failure of the sensor system due to its large core diameter. Consequently, fibres can be connected on site in industrial environments with relative ease and without affecting the system. Plastic optical fibres are also considered easier to handle when compared to glass optical fibres. Glass fibres tend to break when bent around a small radius, which does not occur with plastic fibres. These fibres are extremely low in cost when compared with glass optical fibres. [2]

Within the radiation industry, dosimetry techniques involving the use of dyed PMMA (poly methylmethacrylate) slabs is one of the most widely used, due primarily to its reliability and stability. These PMMA slabs change colour as they are exposed to radiation and this radiation-induced attenuation is monitored using a spectrophotometer post-irradiation. Different ranges can be monitored depending on the dye used, e.g., Harwell Red 4034 has a dose range of 5 kGy to 50 kGy, and Harwell Gammachrome YR has a dose range of 100 Gy to 3 kGy [3]. One of the main drawbacks of this system however, is the lack of real-time information. The PMMA slabs must be removed from the area of interest to be tested in a laboratory, and thus there is no information available on the dose until after irradiation is completed.

Based on the same principle of radiation-induced attenuation used by commercial PMMA slabs, an in-situ system based on PMMA polymer optical fibres can be realised giving real-time information of the radiation dose received.

II. APPLICATIONS OF IONISING RADIATION

One of the main application areas of gamma radiation is in sterilisation, in particular, the sterilisation of medical products using gamma radiation. The ability of radiation to kill pathogenic micro-organisms is the basis on which the sterilisation of medical products depends. It is used in a wide variety of products such as hypodermic needles, surgical sutures, blood handling equipment, implant substances and tissues, surgical gloves and utensils, catheters, dental supplies, etc. It is still unknown how exactly the radiation kills micro-organisms, however it is thought to be associated with the damage caused by the radiation to the deoxyribonucleic acid (DNA) of the micro-organism [4]. The radiation sensitivity of micro-organisms depends on the amount of DNA in the nucleus, but it also depends on a number of other factors, such as the environment they are in. Micro-organisms irradiated in an aerobic environment are more sensitive than those in an anaerobic environment. Also, those in water are more sensitive than those in the dry state. The exact dose of gamma radiation used in the sterilisation of medical products varies for different countries but is usually between 25 kGy and 35 kGy [4].

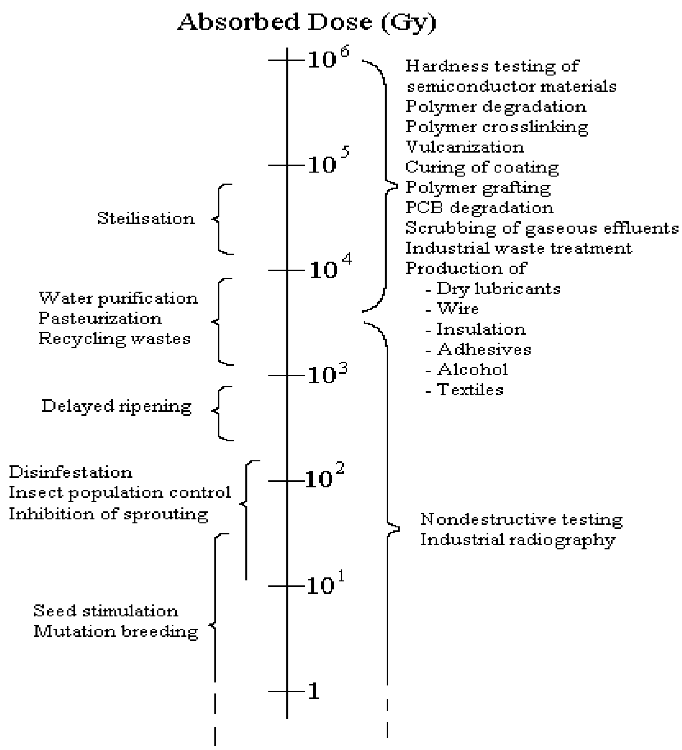


Figure 1: Dose ranges for various applications of ionizing radiation [4]

Gamma radiation can be used for a number of different applications within the food industry. Food can be treated to prevent sprouting in onions, garlic and potatoes, to extend the shelf-life of mushrooms, cherries and strawberries, to eradicate insects in grain and fruit, to kill pathogenic microorganisms in fish and meat, to pasteurize dried herbs and spices, and to delay ripening of fruit and vegetables. Radiation can also be used to prevent the spoilage process, which commonly leads to the rotting of food and wastage. This is due to radiation slowing down the physiological, chemical and biochemical changes occurring in the food and killing micro-organisms and insects. Although radiation does not prevent the drying out of fresh food, its application allows the food to be treated in a sealed package, which prevents both the desiccation process and the microbial re-contamination. Food irradiation treatment depends on the ability of radiation to kill cells and alter the enzyme activities of the food. The killing action of radiation, which inhibits cell division, is used to prevent sprouting, reduce the number of viable micro-organisms on the food, prevent the hatching of insect eggs and larvae, and kill or sterilize the insects in the food. A radiation treatment of between 1 kGy to 7 kGy, depending on the product, can significantly reduce the number of viable pathogenic micro-organisms, e.g. *Salmonella*, *Escherichia coli* and *Lysteria*, which can contaminate food causing serious food poisoning [4].

A number of other applications for radiation are also being investigated, such as in the treatment of sludge, to reduce the amount of bacteria and infectious micro-organisms, before it may be used as a fertiliser. It is also used in the preservation of ancient objects having historic or artistic value by killing the micro-organisms that can destroy the organic material that the objects are made of or have components of. [4] A summary of some of the different application of radiation is shown in figure 1, along with the absorbed doses for the specific process.

III. PROPERTIES OF RADIATION DOSIMETERS

In all the radiation processes, dosimetry is involved, as a necessary control, to establish the processes or occasionally, for research and development studies. Dosimetry provides the quantitative baseline against which the biological or chemical changes induced by the radiation can be measured. It is, therefore, the accepted methodology that is applied to ensure that a radiation process meets the relevant specifications and also to ensure that results obtained in the laboratory can be reproduced elsewhere, either in other laboratories or in a commercial radiation facility.

A number of physical and chemical sensors, which can be subdivided into liquid, solid and gaseous systems, are available to measure ionising radiation and researchers are continually looking for ways to improve the systems, be it increasing the sensitivity, providing real-time measurements or significantly reducing the costs. All these factors are important for providing the optimum radiation dosimeter [5].

Material sensitivity: The ability for radiation to interact with the material is the primary characteristic for any radiation dosimeter. This interaction varies greatly among the different types of radiation dosimeters. The materials utilised in dosimetry must have a high sensitivity to the ionising radiation within the dose ranges required for the specific application.

Post-irradiation fading: The material used as a radiation dosimeter must also show minimum post-irradiation fading. This post-irradiation fading is due to the repair of the physical damage caused to the material during irradiation. Information regarding the irradiation dose is lost if the dosimeter material begins to recover immediately after irradiation.

Time dependence: Many materials also exhibit a time dependence of specific absorbance. This means that it is often required to wait up to 3h before obtaining an accurate reading. For medical applications it is usually important that the dosimeter is fast to measure.

Stability and reliability: Stability and reliability of the dosimeter, as with any sensor, are also important. In order to ensure that this can be achieved the dosimeter must be immune to a number of environmental conditions. The effect of humidity and temperature on the dosimeter must be investigated and accounted for. The influence of dose rate on the dosimeter material should also be considered. Immunity to other disturbances, such as those found in electromagnetically harsh environments, is also advantageous for gamma dosimeters as they are often employed in such conditions.

Ease of use: For all applications it is important that the dosimeters are easy to use. For this to be achieved, the system must be easily installed in the area of application without the risk of affecting the measurements. Maintenance should also be minimal. The readout from the dosimeter should be clear and easy to understand. It is also important that the system is low in cost.

IV. PMMA RADIATION DEGRADATION

The degradation of PMMA due to exposure from ionizing radiation has been investigated in depth by Yoshida and Ichikawa [6]. In general, the effects of ionizing radiation can be divided into two sections; i) main-chain scission (degradation) and ii) crosslinking. In many polymers both processes take place in parallel with one another, however in certain cases the scission predominates the crosslinking, and such polymers are known as degrading polymers. PMMA, shown in figure 2, is one such polymer.

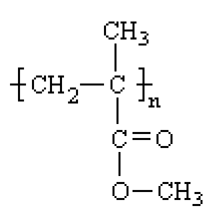
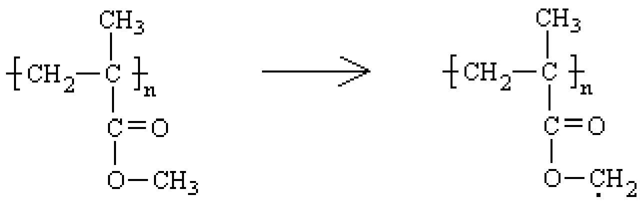


Figure 2: PMMA polymer molecule

Yoshida and Ichiwkawa [6] report that the side-chain is initially affected by the gamma irradiation and the radical formed is a precursor for the main chain scission. When PMMA is irradiated with ionizing radiation, such as gamma radiation, a free radical is generated on the ester side-chain, $-COO\dot{C}H_2$.



Equation 1

This side chain radical may be generated in a number of ways [7];
by direct action of ionizing radiation:

$$-COOCH_3 + \gamma \rightarrow -COO\dot{C}H_2 + H \qquad \textbf{Equation 2}$$

by proton transfer of the side-chain cation:

$$\begin{gathered} -COOCH_3 + \gamma \rightarrow -COOCH_3^{\cdot +} + e^- \\ -COOCH_3^{\cdot +} \rightarrow -COO\dot{C}H_2 + H \end{gathered} \qquad \textbf{Equation 3}$$

or by hydrogen abstraction:

$$-COOCH_3 + H \rightarrow -COO\dot{C}H_2 + H_2 \qquad \textbf{Equation 4}$$

A) Radiation – Induced Attenuation of PMMA optical fibres

The Beer-Lambert Law can also be used to determine the radiation dose absorbed. The absorption co-efficient, α, is given by equation 5.

$$\alpha = -\frac{1}{L_0}\log\left\{\frac{P(\lambda)}{P^0(\lambda)}\right\} \qquad \textbf{Equation 5}$$

where $\frac{P(\lambda)}{P^0(\lambda)}$ is the ratio of the spectral radiant power of light transmitted through the irradiated dosimeter to that of the light transmitted in the absence of the dosimeter.

The Beer-Lambert Law is used to determine the radiation-induced attenuation. The radiation-induced attenuation is given by equation 6 and can be further explained through the diagram in figure 3.

$$RIA(dB) = -\frac{10}{L_0}\log\left\{\frac{P_T(\lambda,t)}{P_T^0(\lambda)}\right\} \qquad \textbf{Equation 6}$$

where L^0 is the irradiated length of fibre, $P_T(\lambda,t)$ is the measured optical power in the irradiated fibre and $P_T^0(\lambda)$ is the optical power of the reference fibre.

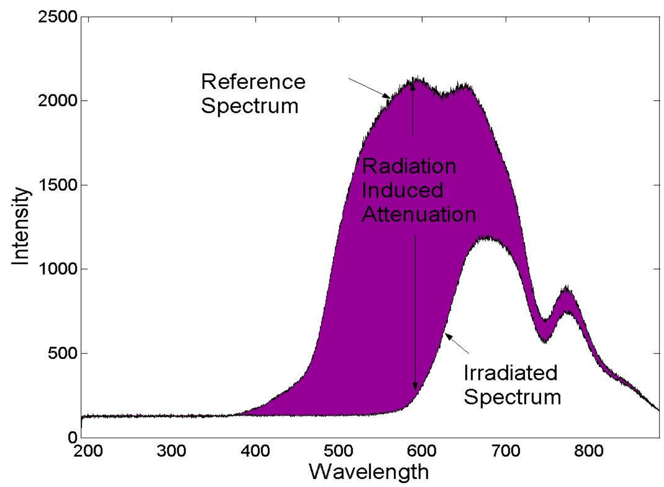


Figure 3: Diagram showing how radiation-induced attenuation measurements are obtained from the transmission spectra.

V. EXPERIMENTAL SET-UP

Radiation measurements were performed at the Isotron gamma radiation facility in County Mayo, Ireland. A 7m length of PMMA based optical fibre was placed in the irradiation chamber, where it was irradiated by the ^{60}Co source, at a dose rate of 25kGy/hr. Access to the irradiation chamber is through a shielded maze and so two 50m length of fibre, allowed for remote monitoring of the radiation-induced attenuation by transmitting the optical signal from a deuterium halogen white light source through the PMMA fibre in the irradiation chamber and returning to an OceanOptics S2000 Dual-Channel spectrometer within the control area. The PMMA based plastic optical fibres used in all experiments were supplied by Fibre Data and consisted of a 1mm PMMA core, fluorinated PMMA cladding and polyethylene jacket, (ncore ~ 1.49, attenuation ~ 0.15 dB/m), terminated with SMA connectors.

VI. RESULTS AND DISCUSSION

The transmission spectra were recorded in realtime and analysis done is on individual wavelengths. Figure 4 shows the transmission spectrum before the fibre were irradiated and after it was irradiated to 3kGy of gamma radiation. It is immediately clear that, after exposure to gamma radiation, the transmission is attenuated over the entire visible spectrum. The results show that the amount of attenuation depends on the wavelength, with lower wavelengths having a significantly higher attenuation than higher wavelengths.

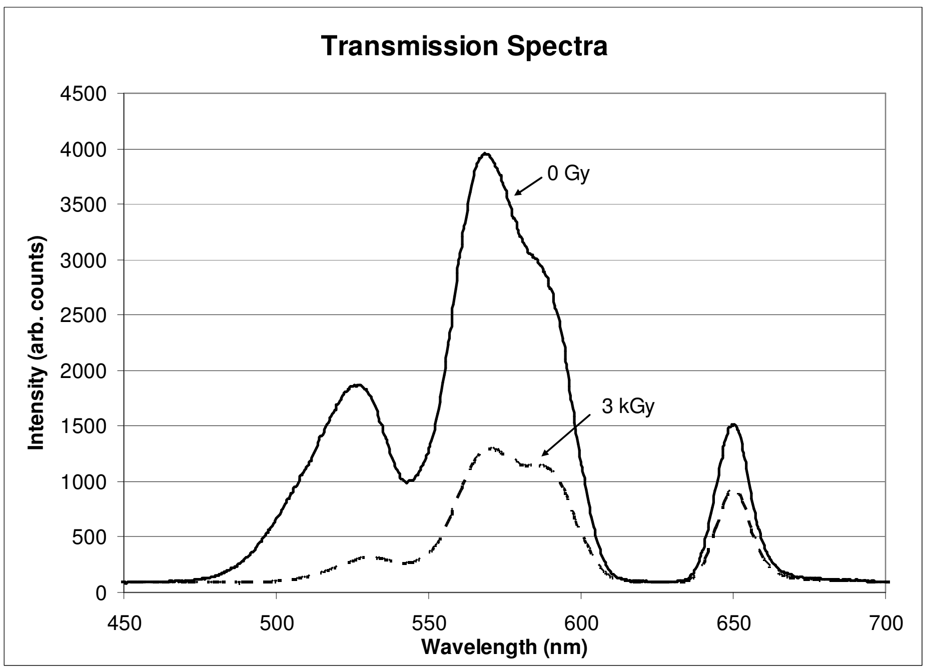


Figure 4: Transmission spectra before irradiation and after 3kGy.

The peak wavelengths were selected for further analysis and are presented in figure 5. It can be seen that as the wavelengths increase, the rate at which the intensity decreases is reduced. 525nm shows a rapid decrease in intensity and becomes fully attenuated rapidly. In contrast to this, at 650nm the intensity, while already low to begin with due to the intrinsically high attenuation in PMMA in this region, decreases at a much slower rate and does not become completely attenuated over the course of the experiment.

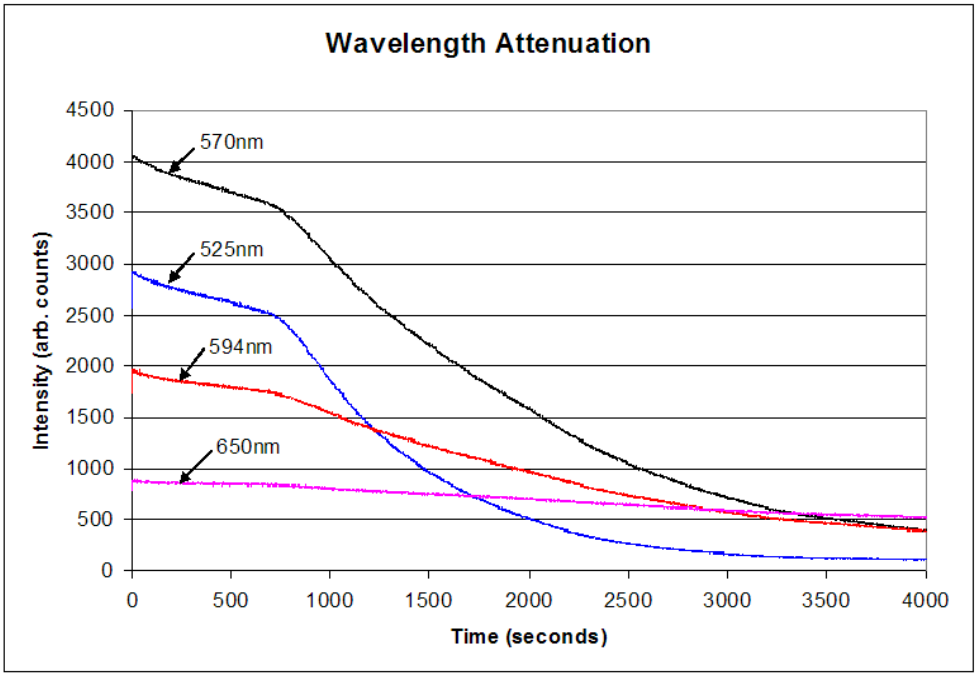


Figure 5: Intensity of the peak wavelengths over time, as they are exposed to gamma radiation.

The sensitivity of the PMMA fibre to ionizing radiation is directly related to the wavelength and thus by selecting the correct wavelength the fibre can be used to monitor over a wide dose range, selecting 525nm for low doses with high sensitivity and selecting 650nm for high doses with lower sensitivity.

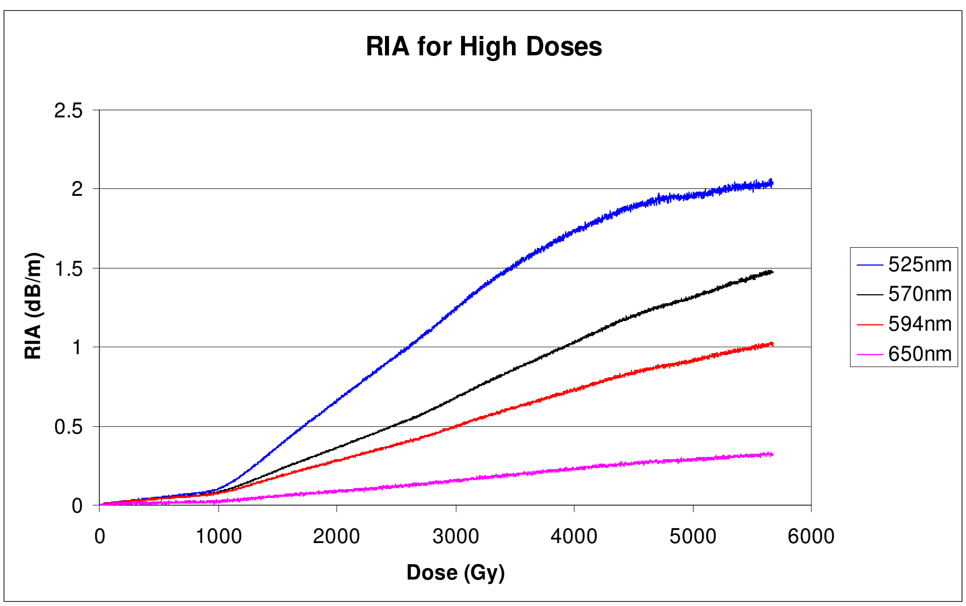


Figure 6: Radiation-induced attenuation for the peak wavelengths.

Radiation-induced attenuation (RIA) calculations, based on equation 6, were performed on the individual wavelengths to determine the exact sensitivity of the fibre to gamma radiation and the results are presented in figure 6. The wavelength dependence on the RIA sensitivity is evident as the rate of attenuation decreases for increasing wavelength. All the wavelengths exhibit identical characteristics in the RIA with increasing dose. Initially, up to 1kGy, the RIA increases slowly, followed by significantly higher rate of increase. After this the fibre reaches saturation, whereby the intensity has become completely at that wavelength. The saturation can be seen in figure 7, and indicates the dose range over which the individual wavelengths can monitor. As the wavelength increases the upper-detection limit for that individual wavelength also increases, 4kGy at 525nm and 45kGy at 650nm.

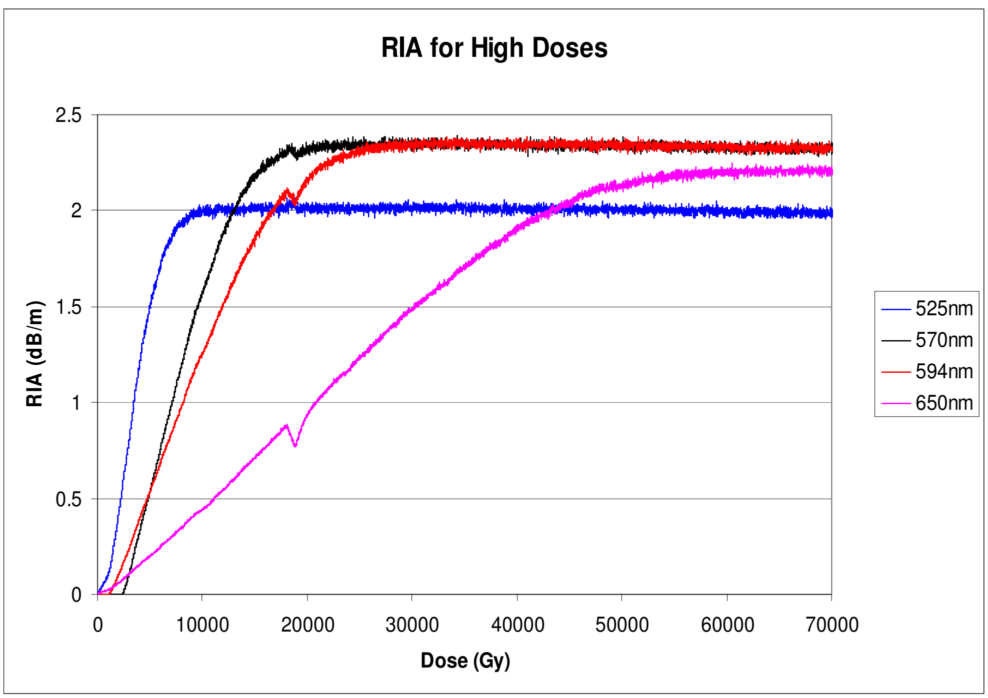


Figure 7: RIA for high doses of gamma radiation.

Figure 8 shows the radiation-induced attenuation at lower radiation doses up to 50Gy for the two lower peak wavelengths, 525nm and 570nm. The determination of lower detection limit of the fibre was limited by the experimental set-up and the high dose rate of the gamma source used, however it can be seen that there is a distinct and immediate increase in the radiation-induced attenuation at these wavelengths. An averaging filter is applied to the data and indicates that beyond 30Gy there is a steady, quantifiable increase in the radiation-induced attenuation.

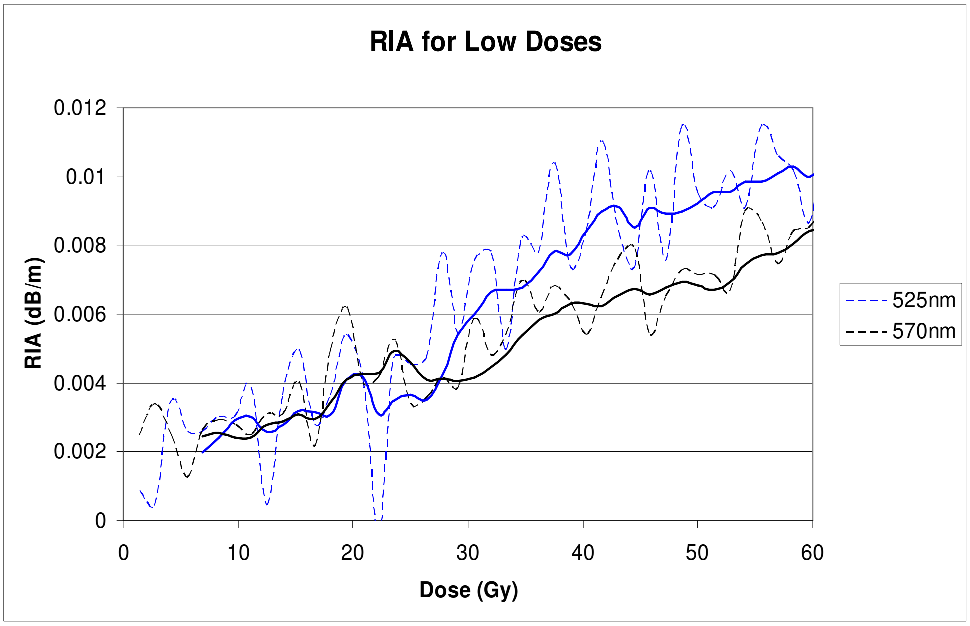


Figure 8: RIA for low doses of gamma radiation.

Table 1 gives a summary of the sensitivity and dose range of the PMMA fibre for individual wavelgnths. It shows that the sensitivity decreases with increasing wavelengths, from 0.6dBm-1/kGy at 525nm to 0.06dBm-1/kGy at 650nm. The dose range is also dependant on the wavelength as those wavelengths with high sensitivity becoming completely attenuated relatively quickly compared with the higher wavelengths.

Wavelength (nm)	Sensitivity (dBm^{-1}/kGy)	Dosimetry range (kGy)
525	0. 6	0.03 - 4
570	0. 3	0.03 - 10
594	0. 2	0.5 - 12
650	0.06	1 - 45

Table 1: Summary of wavelength dependant sensitivity and dose range

VII. CONCLUSIONS

The use of PMMA based polymer optical fibres has been demonstrated to be effective in monitoring gamma radiation over a wide dose range. PMMA optical fibres are known for their high attenuation outside of the visible region, however the wavelengths of interest for these measurements lie between 500nm and 700nm, where there is a high optical signal. The sensitivity of the fibres to the radiation dose was seen to be wavelength-dependant, with sensitivity decreasing as the wavelength increases. Due to this variance in sensitivity, it was also found that the dose range was also wavelength dependant, as the fibre was completely attenuated at lower wavelengths while at higher wavelengths, where RIA sensitivity was less, there was still a measurable signal. The fibres exhibit good sensitivity, as high as $0.6dBm^{-1}/kGy$, and are capable of monitoring dose ranges between 30Gy and 45kGy. This exceeds the sensing range of all currently available sensors. The dose range can also be improved by altering the length of fibre used. Increasing the length of fibre irradiated will allow for higher sensitivity at low dose ranges, while by using shorter lengths of fibres with a higher optical power source, higher dose monitoring limits can be attained. The exact sensitivity and dose range can be chosen by careful selection of the monitoring wavelength, thereby allowing the PMMA fibre to be used for a number of different applications within the radiation processing industries.

VIII. ACKNOWLEDGEMENTS

The authors would like to thank the staff of Isotron Ireland for their help and use of facilities, and also John Cronin for his assistance in these experiments. This research was funded under the by the European Commission under the 7th Framework Programme through the 'Marie Curie Re-integration' action of the 'Peoples' Programme, (PERG04-2008-239207).

IX. REFERENCES

1. Ziemann, O., et al., *POF Handbook - Optical Short Range Transmission Systems*. 2nd ed. 2008, Berlin: Springer.
2. Weinert, A., *Plastic Optical Fibers*, ed. B.a.M. Siemens Aktiengesellschaft. 1999: Publicis MCD Verlag.
3. Harwell Dosimeters Ltd. *http://www.harwell-dosimeters.co.uk*. [cited 2005 25 July 2005]; Available from: http://www.harwell-dosimeters.co.uk.
4. McLaughlin, W.L., et al., *Dosimetry for radiation processes*. 1989: Taylor & Francis. 251.
5. O'Keeffe, S., et al., *A Review of Optical Fibre Radiation Dosimeters,* Sensor Review, 2008. **28**(2): p. 136-142.
6. Yoshida, H. and T. Ichikawa, *Temperature effect on the radiation-degradation of poly(methyl methacrylate).* Radiation Physics and Chemistry, 1995. **46**(4-6, Part 1): p. 921.

7. Ichikawa, T., *Mechanism of radiation-induced degradation of poly(methyl methacrylate) -- temperature effect.* Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 1995. **105**(1-4): p. 150.