

Photon dosimetry using Red 4034 Harwell dosimeters in a pool-type research reactor

J.P. Santos^{a,*}, J.G. Marques^{a,b}

^a*Instituto Tecnológico e Nuclear, Estrada Nacional 10, 2683-953 Sacavém, Portugal*

^b*Centro de Física Nuclear da Universidade de Lisboa, Av. Prof. Gama Pinto, 2, 1649-003 Lisboa, Portugal*

Abstract

The performance of Red 4034 Perspex dosimeters from Harwell Technologies in routine photon dosimetry in mixed fields was evaluated. The dosimeters were irradiated in core grid positions of the Portuguese Research Reactor under a photon dose rate of the order of 10^4 Gy/h, a thermal neutron flux in the range of 1.8×10^{10} to 1.1×10^{11} n/cm²/s and temperatures below 40 °C. The dosimeters visibly darken upon irradiation and this effect, accurately measurable by spectrophotometry, is related to absorbed dose. Thermoluminescence dosimeters and a calibrated ionization chamber were used as comparison. The results show that Red 4034 Perspex is a suitable dosimeter for mixed-field dosimetry up to 50 kGy.

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1. Introduction

Radiation fields in fission reactors are complex and the different components must be discriminated. While the characterization of the neutron field can be performed using activation foils, the measurement of the photon dose rate normally requires the use of costly ionization chambers.

The aim of this work is to evaluate the performance of Red 4034 Perspex dosimeters from Harwell Technologies (UK) for routine photon dosimetry in the mixed radiation field of the Portuguese Research Reactor (RPI), at the Nuclear and Technological Institute (ITN, Portugal).

The RPI is a pool-type, natural water moderated and cooled reactor, with 1 MW power. The activities currently underway cover a broad range of applications from irradiation of electronic circuits (Franco et al., 2005) to calibration of detectors for dark matter search (Girard et al., 2005), through more classical subjects such as neutron activation analysis (Dias and Prudêncio, 2007). Each activity has specific dosimetry needs.

Red 4034 dosimeters were originally developed in the 1960s and have shown to be suitable to monitor doses during the radiation sterilization of medical products or during the radiation pasteurization of food products, in pure photon fields at room temperature (Whittaker et al., 1970).

In this work the measurement capabilities of Red 4034 dosimeters were evaluated for photon dosimetry in mixed photon–neutron fields at temperatures in the 30–40 °C range. The results obtained with the Red 4034 measurements are compared with results obtained with thermoluminescence (TL) dosimeters and with an ionization chamber in the same experimental conditions.

2. Materials and methods

Polymethylmethacrylate (PMMA) dosimeters were developed at Harwell for high photon doses measurements in industrial radiation processing (ISO/ASTM 51276, 2006). The dosimeters are machined in 30 × 11 mm rectangles with thickness 3.00 ± 0.55 mm (Harwell Dosimeters, 2007) from Red 4034 Perspex sheets and sealed in individual polyester/aluminium foil/polyethylene laminate sachets.

* Corresponding author.

E-mail address: joanas@itn.pt (J.P. Santos).

When exposed to ionizing doses above 1 kGy, Red 4034 starts to darken due to the formation of a new absorption band peaking at 615 nm and extending from 600 nm to beyond 700 nm. The absorbed dose is determined by measuring its radiation-induced absorbance in the 630–650 nm range where post-irradiation fading is minimal (Whittaker et al., 1970), thus 640 nm is commonly used as readout wavelength.

The specific absorbance (absorbance/cm) is usually converted to the derived dose using the calibration curves supplied by the manufacturer for each specific batch. When the dosimeters are exposed to a wide range of dose rates and temperatures and when they are exposed to high temperatures for several hours after irradiation and before measurement it is advisable to do a calibration under the same irradiation conditions of the experiment (Fairand, 1998).

The main factors that affect the dosimeter photon response and stability are the water concentration in the PMMA gained during its production, the photon dose rate and the temperature during and after an irradiation (Barrett et al., 1990; Biramontri et al., 1996; Whittaker et al., 2001).

The packaging protects the PMMA from surface damage that could affect the absorbance measurement accuracy and preserve the optimized level of water concentration achieved at the dosimeter manufacture process (Barrett et al., 1990). Being thus, the dosimeters were irradiated in their sealed sachets and only removed shortly before readout.

Red 4034 are essentially temperature-independent up to 40 °C if the readout is performed within the 2 days after irradiation (Barrett et al., 1990). However, some studies revealed that temperature effects are batch-dependent, slightly at 30–40 °C but larger for higher temperatures up to 50 °C (Whittaker et al., 1985).

The effect of post-irradiation time on the dosimeter response depends on water concentration, temperature, dose, dose rate and readout wavelength (Bett et al., 2002). Effects of postirradiation temperature are significant above 40 °C affecting the specific absorbance from $\pm 3\%$ to $\pm 12\%$ depending on batch and dose rate (Whittaker et al., 2001). When the irradiated dosimeters are stored at normal ambient temperatures (15–25 °C) the specific absorbance falls $\pm 2\%$ over 10 days (Whittaker et al., 1985). Readout within two days after the irradiation is not necessarily preferable to readout within a longer interval when the dose rate is higher than 15 kGy/h (Fernandez Fernandez et al., 2005).

Low dose effects appeared at rates below 5.4 kGy/h and specially if combined with higher temperatures (Whittaker et al., 1985). At dose rates above 5.4 kGy/h and temperatures in the range of 20–40 °C Red 4034 Perspex performs quite well in pure photon fields (Glover, 1993; Fernandez Fernandez et al., 2005).

Red 4034 Perspex dosimeters from a “JB” batch were used in the measurements performed at RPI. Dosimeters from this batch were calibrated in the dose range of 5–50 kGy, under a photon dose rate of 5.3 kGy/h, using the ^{60}Co source from the Radiation Technology Unit (UTR) at ITN. Ceric-cerous was selected as the reference standard dosimeter (ISO, 1995) for the local calibration of the ^{60}Co source. The Red 4034 dosimeters

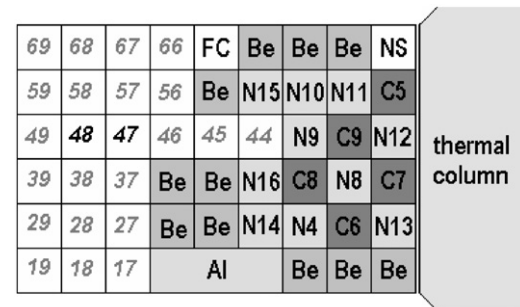


Fig. 1. RPI N2-P1/9 configuration: Be—beryllium reflector; Al—aluminium reflector; C—control fuel element; N—standard fuel element; FC—fission chamber; NS—neutron source. Measurements were performed in the grid positions labelled in bold italic.

were analyzed with a Shimadzu UV mini-1240 spectrophotometer, with the absorbance being measured at 640 nm. The thickness of each dosimeter was measured with a calibrated Mitutoyo dial gauge with an accuracy of ± 0.01 mm.

Mixed-field irradiations were performed in free core grid positions in the reactor pool. The reactor core is mounted on a 6×9 grid plate, shown in Fig. 1, with a $77.0 \text{ mm} \times 81.0 \text{ mm}$ pitch. The free positions (labelled in italic) can be used for irradiations.

The Red 4034 dosimeters were irradiated in grid positions 47 and 48 at 100 kW in order to maintain an air-kerma rate of 10^4 Gy/h and a thermal neutron flux in the range of 1.8×10^{10} to $1.1 \times 10^{11} \text{ n/cm}^2/\text{s}$. The temperature at the location of the dosimeters was below 40 °C. The distance between the centre of the two irradiation positions is 81.0 mm, as defined by the core grid.

The dosimeters were placed in polyethylene holders covering about 15 cm of the mid-height of the fuel elements. One Al activation foil was placed behind the Red 4034 located at the mid-height of the fuel elements to control the experimental conditions. A dose value is achieved from the average reading of the three Red 4034 dosimeters.

A previous characterization of the radiation field in grid positions in the same experimental conditions has been done with aluminium oxide (Al_2O_3 :Mg, Y) TL dosimeters (type D-2, Institute of Isotopes and Surface Chemistry, Hungary) (Osvay et al., 1993), and with a neutron-insensitivity closed stainless steel ionisation chamber filled with nitrogen (type CRGA 11, Radiotechnique Compelec, France). The ionization chamber has a sensitivity of $4.28 \times 10^{-7} \text{ A/Gy/s}$ for ^{60}Co in air and a good accuracy (5–6%) for photon dose measurements in mixed fields. The D-2 dosimeters have proved to be suitable for photon dosimetry in mixed fields not only because of their linearity over a wide dose range (1 mGy–10 kGy) (Santos et al., 2007) but also for their lower sensitivity (3.4 mGy per 10^{10} n/cm^2) to thermal and fast neutrons up to 4 MeV (Osvay, 1996).

3. Results and discussion

Fig. 2 shows the specific absorbance at 640 nm as a function of the photon dose and compares the calibration curve provided by Harwell with the one obtained from the UTR irradiations.

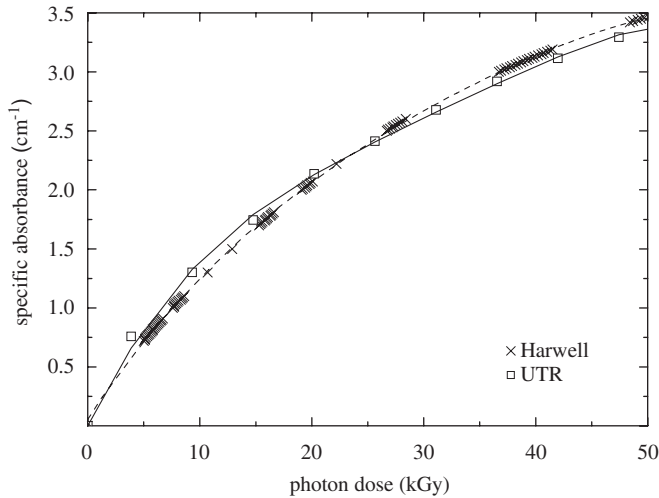


Fig. 2. Specific absorbance versus photon dose.

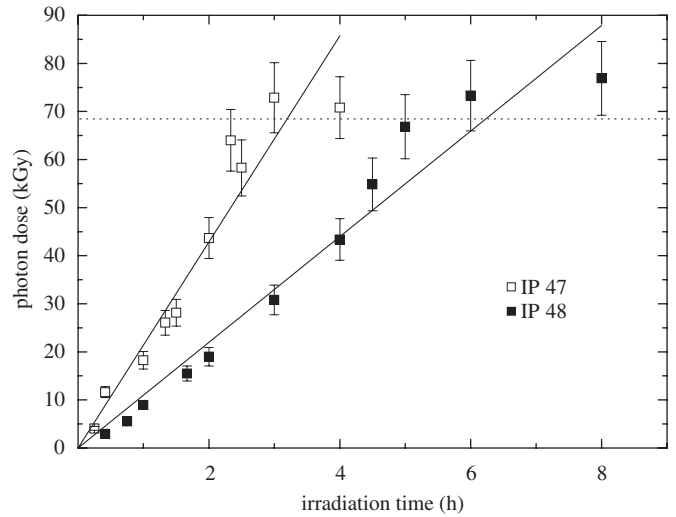


Fig. 3. Photon dose measurements in two grid positions (IP47 and IP48: irradiation positions at 283.5 and 365.5 mm from the face of the core, respectively) at different irradiation times. The error bars correspond to a 10% uncertainty.

Table 1
Fading of Red 4034 Perspex irradiated at different doses levels at RPI

Photon dose (kGy)	Elapsed time (days)					
	1–2%	7%	8%	12%	14%	22%
> 50		95.5	94.8	95.5	95.5	–
48.1		95.5	95.5	95.5	95.5	–
23.0	100	100	100	95.5	95.5	95.5
11.0		100	100	100	95.5	95.5
6.0		100	100	100	95.5	95.5

Table 2
Photon dose rate obtained with three different methods in core grid positions at 100 kW

Core grid positions	Photon dose rate (kGy/h)		
	Red 4034 (640nm)	D-2	CRGA11
47	21.43 ± 2.14	22.98 ± 0.93	22.81 ± 1.28
48	10.98 ± 1.10	11.20 ± 0.45	12.11 ± 0.68

The maximum deviation is of the order of 10% relative to the calibration provided by the manufacturer and confirms the importance of using a specific calibration for the instrument in use.

The fading characteristics of Red 4034 dosimeters irradiated at the RPI for different dose levels were studied for a period of 22 days. After each readout the dosimeters were re-sealed into sachets. The results are summarized in Table 1. Fading was found to be more significant at high doses and is shown to be negligible over the first few days after the irradiation. The results presented here were obtained reading the dosimeters within two days after the irradiation.

Fig. 3 shows the results obtained in several irradiations in grid positions 47 and 48. Red 4034 dosimeters show a linear dose response up to about 50 kGy, with a tendency to saturate after this value. The corresponding neutron fluences are approximately 3×10^{14} n/cm² (thermal, < 0.5 eV), 3×10^{13} n/cm² (fast, > 1 MeV) for irradiation position 48 and to 1×10^{15} n/cm² (thermal, < 0.5 eV), 3×10^{14} n/cm² (fast, > 1 MeV) for irradiation position 47. The determined photon dose rates are 10.98 kGy/h (position 48) and 21.43 kGy/h (position 47).

The uncertainty value in the measurements performed with the Red 4034 Perspex was estimated to be ±10%, taking into account the uncertainty in the absorbed doses reported by Harwell, the uncertainty in the spectrometric analysis of dosimeter response and measurement of dosimeter thickness;

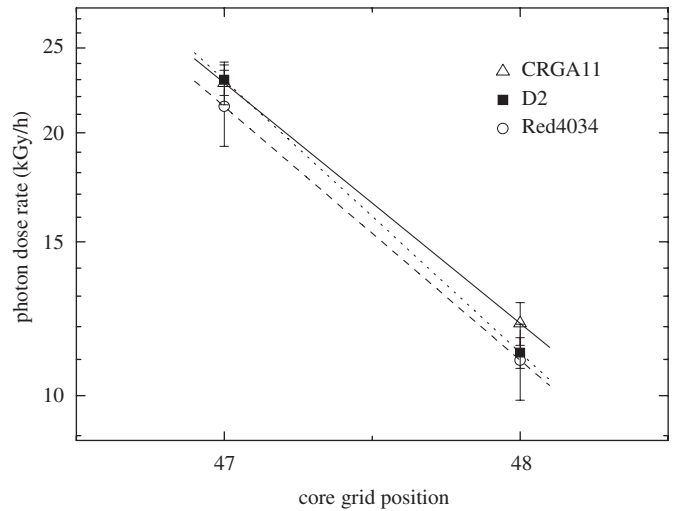


Fig. 4. Photon attenuation from the face of the core for positions 47 and 48.

goodness of fit function to the calibration data and an uncertainty associated with the irradiation conditions.

The reproducibility of the measurements performed with Red 4034 dosimeters was studied in position 48 at a reactor power of 100 kW, in irradiations of 2 h and in different days and it was found to be better than 7%. This includes already the uncertainties in reactor power.

Table 2 summarizes the results obtained at 100 kW with Red 4034 dosimeters, D-2 dosimeters and with the ionization chamber (CRGA11) and Fig. 4 shows the variation of the radiation field from the face of the core for 283.5 and 364.5 mm.

The photon dose rates obtained with the three methods are in agreement within 10%, which is sufficient for most radiation processing applications.

4. Conclusions

Red 4034 performs relatively well in mixed fields, having determined photon doses with good linearity up to 50 kGy. A comparison of the three methods applied to determine the photon dose in the mixed fields of the open pool-type research reactor shows an agreement within 10%, which is acceptable for the intended application.

Red 4034 dosimeters have same advantageous properties like the simplicity of handling and low price, allowing the positioning of the dosimeters near the materials to be irradiated and should extend the range of photon dose measurements up to 50 kGy.

Experiments at powers above a few tenths of MW cannot be directly monitored by Red 4034, but the characterization of the irradiation position at full power can be done at a lower power, e.g., 100 kW, as used here and the results extrapolated. An ^{16}N linear channel (IAEA, 1997), available in the instrumentation and control system of many research reactors can minimize errors in the scaling of the values. This channel does not suffer from the small fluctuations that the neutron channels might show (connected with the ^{135}Xe cycle, especially if the reactor has an irregular operating schedule) and has a wider linear range than a thermal channel (measuring the temperature increase of the coolant across the core).

The study of the photon dose measurements reproducibility performed with Red 4034 is being extended to different experimental conditions and a study of the influence of the packaging will also be performed with the goal of improving the holders for positioning the dosimeters in the core grid.

Further work will include the evaluation of the advantages of using the PMMA dosimeters in the calibration of TL dosimeters for dose levels up to 10 kGy (saturation value of Al_2O_3 D-2 type) at positions with low neutron–photon ratios.

The performance of Red 4034 dosimeters to monitor photon doses will also be extended to the fast neutron irradiation facility for electronic components of the RPI (Franco et al., 2005).

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