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Neutron dose estimation in a zero power nuclear reactor

S. Triviño [a,b,*], J. Vedelago [a,c], F. Cantargi [d], W. Keil [b,g], R. Figueroa [e,h], F. Mattea [a,f], A. Chautemps [b], M. Santibañez [e,h], M. Valente [a,c,e,h,**]

[a] Laboratorio de Investigación e Instrumentación en Física Aplicada a la Medicina e Imágenes por Rayos X (LIIFAMIRx), Universidad Nacional de Córdoba, Av. M. Allende s/n, Córdoba, Argentina
[b] Reactor Nuclear RA-0, Comisión Nacional de Energía Atómica, Av. Vélez Sarsfield 1611, Córdoba, Argentina
[c] Instituto de Física Enrique Gaviola (IFEG) - CONICET, Av. M. Allende s/n, Córdoba, Argentina
[d] Centro Atómico Bariloche, Comisión Nacional de Energía Atómica, Av E. Bustillo km 9500, Bariloche, Argentina
[e] Departamento de Ciencias Físicas, Universidad de La Frontera, Francisco Salazar 1145, Temuco, Chile
[f] Facultad de Ciencias Químicas, Universidad Nacional de Córdoba, Haya de la Torre s/n, Córdoba, Argentina
[g] Facultad de Ciencias Exactas, Físicas y Naturales, Universidad Nacional de Córdoba, Av. Vélez Sarsfield 1611, Córdoba, Argentina
[h] Centro de Física e Ingeniería en Medicina (CFIM), Universidad de la Frontera, Francisco Salazar 1145, Temuco, Chile

HIGHLIGHTS

- Dose components in a neutron mixed field were characterized using Fricke dosimetry.
- Dedicated MCNP routine were developed to perform dosimetry in RA-0 mixed field.
- Results were successfully compared with activation foils measurements.

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ABSTRACT

This work presents the characterization and contribution of neutron and gamma components to the absorbed dose in a zero power nuclear reactor. A dosimetric method based on Fricke gel was implemented to evaluate the separation between dose components in the mixed field. The validation of this proposed method was performed by means of direct measurements of neutron flux in different positions using Au and Mg-Ni activation foils. Monte Carlo simulations were conversely performed using the MCNP main code with a dedicated subroutine to incorporate the exact complete geometry of the nuclear reactor facility. Once nuclear fuel elements were defined, the simulations computed the different contributions to the absorbed dose in specific positions inside the core. Thermal/epithermal contributions of absorbed dose were assessed by means of Fricke gel dosimetry using different isotopic compositions aimed at modifying the sensitivity of the dosimeter for specific dose components. Clear distinctions between gamma and neutron capture dose were obtained. Both Monte Carlo simulations and experimental results provided reliable estimations about neutron flux rate as well as dose rate during the reactor operation. Simulations and experimental results are in good agreement in every positions measured and simulated in the core.

1. Introduction

Neutron dosimetry is a very important task in several fields such as medical purposes (Blaumann et al., 2004; Schütz et al., 2011), nuclear research laboratories (Harling et al., 2005), nuclear power plants (Wolber et al., 1996) and industry (Rodriguez-Carvajal, 1993).

After successive collisions, neutrons are thermalized due to the energy losses associated with the interactions. Thermal neutrons continue their propagation in matter until they are captured by an atomic nucleus. Some of the reactions that can take place with light atomic number organic compounds are: $^{1}H(n, \gamma)^{2}H$, $^{14}N(n, p^{+})^{14}C$, $^{14}N(n, \gamma)^{15}N$, $^{16}O(n, \gamma)^{17}O$, and other reactions involving Cl, Na, P, S, K, and Ca nuclei.

* Corresponding autor at: Reactor Nuclear RA-0, Comisión Nacional de Energía Atómica, Av. Vélez Sarsfield 1611, Córdoba, Argentina.
** Corresponding author.
E-mail addresses: trivino@famaf.unc.edu.ar, trivino.sebastian@gmail.com (S. Triviño), valente@famaf.unc.edu.ar (M. Valente).

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When considering relevant amounts of energy deposition in biological tissues, only the first two of the above are significant; whereas the contribution of the other reactions might be neglected for dosimetric calculations, as required for example in Boron Neutron Capture Therapy (BNCT). Dosimetry for BNCT is, in fact, very complex due to the variety of mechanisms responsible of energy release from neutrons in matter (González and Santa Cruz, 2012; Rogus et al., 1994; Binns et al., 2005).

Different techniques have been developed to characterize a neutron-gamma mixed field, like thermoluminescent dosimeters (Gambarini et al., 2010), nuclear track detector (FNTD) (Sykora and Akselrod, 2010) and gel dosimeters (Gambarini et al., 2000). If a set of dosimeters is meant to be used, they must have different neutron-to-γ-ray sensitivity ratios, in order to be able to distinguish between the dose due to gamma rays and the one due to neutrons. In radiation protection applications it is common to find the use of semiconductors-charged-particle detectors coated with neutron film (McGregor et al., 2003) and thermoluminescent badget dosimeters (Piesch and Burgkhardt, 1985), such as LiF (TLD-100), ^{7}LiF (TLD-700), ^{6}LiF (TLD-600), and CaF_2 (TLD-300). Also, gaseous detectors are widely used (Charpak et al., 2002). Chemical dosimeters are also a reliable alternative method for neutron dose measurements. The most common and relevant dosimeter of this type is the Fricke dosimeter. Fricke solution was originally proposed by Fricke and Morse (1927). During the 80's Gore et al. proposed the implementation of Fricke dosimetry along with magnetic resonance imaging (MRI) techniques to obtain three-dimensional (3D) dosimetry distribution (Gore and Kang, 1984). Then, Appleby et al. (1987) reported that Fricke dosimetry solutions dispersed throughout a gel matrix could be used to obtain 3D spatial dose information using MRI. The dosimetric basis of the Fricke gel relies on the dose dependent transformation of ferrous ions (Fe^{2+}) into ferric ions (Fe^{3+}) (Charpak et al., 2002) and the consequent correlation between the chemical reaction yield with the absorbed dose.

In the 90's, it was reported that the addition of Xylenol Orange (XO) to the standard Fricke solution induced the formation of the complex XO-ferric ions, which modifies the absorption properties of the dosimetric material. This resulted in a visible light absorption peak centered at about 585 nm (Appleby and Leghrouz, 1991) considerably different from the corresponding value of the standard Fricke solution. With this modification, measurements of light transmission with filters around the wavelength of the mentioned absorption peak provided reliable estimations of the absorbed dose (Bero et al., 1999). Moreover, it is also possible to obtain the spatial distribution of the absorbed dose by means of optical imaging of gel dosimeters constructed in thin layers (Gambarini et al., 2011; Valente et al., 2007). Although ferric ion diffusion represents a well-known drawback for Fricke gel dosimetry, this difficulty can be overcome by means of prompt sample analysis (Gambarini et al., 2004, 2006) and by accurate determination of diffusion properties, which helps to establish suitable elapsed times between the irradiation and readout, considering the system stabilization (Vedelago et al., 2014). In this context, elapsed times between sample irradiation and analysis can be reduced by implementing a suitable method for Fricke-XO gel dosimeters (FXD) imaging by means of a portable instrumentation aimed at performing prompt measurements.

In 2000, Gambarini et al. Gambarini et al. (2000) reported the characterization of fast and epithermal neutron beams using thin layers of FXD with the addition of boron-10 (^{10}B) to increase the sensitivity of the gel to neutrons, due to the high cross section of thermal neutron capture. This report presented a possibility of measuring different contributions to the total dose in boron neutron capture treatment using optical techniques. Another independent and reliable method for neutron field characterization is the use of activation foils (Beckurts and Wirtz, 2013; Tripathy et al., 2007). Since photonuclear reactions occur only above the energy (E) range of γ radiation ($E \geq 10$ MeV), metal foils are only activated by neutrons in a mixed n+γ field. Radio-activation methods are used extensively for measuring neutron fluence (Kuijpers et al., 1977). A set of foils with different thresholds can be used to determine the spectrum of neutron flux density. The resulting activity of the foils can be measured with a scintillation counter. The most commonly-used materials are manganese, copper, silver and gold (Beckurts and Wirtz, 2013).

A useful technique for neutron calculation is Monte Carlo simulation. In recent decades, developments in Monte Carlo codes for neutron transport have been significantly improved (Pozzi et al., 2003). In this work, MCNP 5.0-1.6 (Forster et al., 2004) was used. MCNP is a general-purpose Monte Carlo N-Particle code that can be used for neutron, photon, electron, or coupled neutron/photon/electron transport.

This work was carried out in a zero power nuclear research reactor, the *Reactor Nuclear* RA-0, which belongs to the *Comisión Nacional de Energía Atómica* and it is currently placed at the *Universidad Nacional de Córdoba*. This reactor has a maximum allowed power of 1 W. This feature reduces shielding requirements and civil engineering work, as well as the core cooling system requirements and the possibility to access to the reactor core in a short time after its shutdown.

The aim of this work is to characterize the contribution to the total dose of thermal/epithermal neutrons in the core of the RA-0 reactor. The proposed method for distinguishing between different dose components is based on experimental measurements by specific Fricke gel dosimeters and activation foils along with theoretical calculations by means of full stochastic radiation transport implemented in MCNP Monte Carlo simulations including the exact geometric and physical characteristics of the RA-0 reactor.

2. Materials and methods

2.1. Fricke gel dosimeters

Fricke gel dosimeters were prepared using Milli-Q water according to the method described in previous works (Valente et al., 2007). The following concentrations were used: ferrous sulfate [0.6 mM $Fe(NH_4)_2(SO_4)_2$. $6H_2O$], sulfuric acid [250 mM H_2SO_4], porcine skin gelatin 300 bloom [5% of the final weight] as gelling agent and Xylenol Orange [0.28 mM $C_{31}H_{27}N_2$ $Na_5O_{13}S$] as metal ion indicator. In addition to the standard dosimeters, boron (Borax $Na_2B_4O_7$) "^{10}B-doped" dosimeters were made, adding 400 ppm (ppm) of ^{10}B to perform the separation of neutron and gamma dose contributions. The sensitive material was used to fill vials (cuvette type containers of $40 \times 10 \times 10$ mm^3 inner size).

Also a set of the doped Fricke gel dosimeters were irradiated in a conventional X-ray tube available at LIIFAMIR^{x} facilities, using a W anode for calibration purposes. The irradiation was performed with a generator current of 48 mA and voltage of 40 kV at a source to surface distance of 90 cm, resulting in a mean dose rate of (34.8 ± 0.6) Gy/s. A Farmer-type ionization chamber (PTW-Freiburg TN 30013) was used in PMMA phantoms for absolute dose rate measurements.

Before and after irradiation, the transmittance of the Fricke dosimeter cuvettes were measured using a spectrophotometer UNICO, model S1205 Vis, from United Products & Instruments Inc. (USA). Absorption spectra were acquired from 450 nm to 700 nm, with an interval sampling of 1 nm, in order to calculate the integral around the absorption peak.

The component of the absorbed dose due to thermal neutrons (D_{th}) can be assessed by means of the dose values of "standard"

(D_{st}) and ^{10}B-doped (D_{10B}) gel dosimeters according to the expression (1):

$$D_{th} \approx C \cdot [D_{10B} - D_{st}] = C \cdot [\alpha_{10B} \Delta OD_{10B} - \alpha_{st} \Delta OD_{st}] \quad (1)$$

where the coefficient C represents relative sensitivity to boron dose with respect to gamma dose; α_{st} and α_{10B} are the calibration factors (experimentally obtained for each standard and ^{10}B-doped dosimeters batch) that convert optical density differences (ΔOD) in absorbed dose (D). Measurements of light transmission/absorbance were performed by conventional spectrophotometry and ΔOD was calculated integrating around the absorption peak at 540 nm. In a previous work (Gambarini et al., 2000), Monte Carlo simulations have been used to assess the relative sensitivity C values, obtaining that $C \in [0.40, 0.45]$.

Several inaccuracies can be associated to the dose rate determination in Fricke gel dosimeters. Among them, the most relevant ones are the positioning of the cuvettes inside the reactor and the optical analytical method used for the dosimeters read-out. Assuming that the variables are contributing to dose rate errors independently, the overall uncertainty could be obtained as follows:

$$(\sigma_{Fricke})^2 = (\sigma_{position})^2 + (\sigma_{optic})^2$$

where *position* refers to sample positioning inside the irradiation channel in the nuclear reactor; *optic* refers to the optical transmission read-out of the dosimeters and the optical calibration linear fits.

It is worthwhile mentioning that the method described in Eq. (1) is able to calculate the absorbed dose. If damage to biological tissue would be considered, the equivalent dose has to be considered. The equivalent dose (expressed in Sv) can be obtained by multiplying the absorbed dose by the radiation weighting factor. As established by the ICRP 103 (ICRP, 2007), the weighting factor for neutrons is a continuous function of neutron energy (Wrixon, 2008). The main goal in this study is to prove doped Fricke gel dosimeters capabilities for neutron capture dose quantification. Therefore, dose rate estimations were reported in Gy/s units.

2.2. Monte Carlo simulations

Dose and flux due to gamma rays, thermal/epithermal and fast neutrons in the RA-0 nuclear reactor core were calculated using MCNP5 – 1.6. The exact geometry of the reactor was designed according to the structural data of the *Comisión Nacional de Energía Atómica*. Each cell was assigned with its corresponding material.

Inside the reactor core the fuel elements are placed vertically in a circular crown made of three parallel plates of aluminum. Each fuel is a mixture of UO_2, enriched to 20% ^{235}U, graphite and binder pitch, placed inside an aluminum clad with a diameter of 10 mm. The active length of the fuel is 540 mm. It must be mentioned that neutrons are moderated with light water.

The nuclear reactor has three irradiation channels, as shown in green color in the top view of Fig. 1. During the experiments, the dosimeters were located in the irradiation channel at the center of the core, shown in the lateral view of Fig. 1, where the neutron flux is at its maximum.

The central irradiation channel has two different diameters: at the top of the channel the diameter is 35 mm and 460 mm deep; below that point, the diameter is 25 mm. Therefore, in the upper channel two cuvettes were juxtaposed, one filled with standard Fricke and the other one with "^{10}B-doped Fricke". In the lower channel only one dosimeter was placed. Due to the limited dimensions of the bottom part of the irradiation channel, only one vial can be located during irradiation. Thus, requiring the repetition of the experiment in order to get the irradiated pair consisting of the standard dosimeter and the borated dosimeter.

From the simulated neutron flux spectrum, neutron dose rate due to neutron capture by ^{10}B estimation ($\dot{D}_{th}$) was assessed using the differential absorption cross section σ data from ENDF-VI (the same data used by MCNP 5.0 code), according to expression (2):

$$\dot{D}_{th} = k \int \frac{N\varepsilon}{\rho} \varphi_{th}(E)\sigma(E)dE \quad (2)$$

where N is the ^{10}B atom density (expressed in atoms per cm^3), ρ is the mass density of the sample (expressed in kg/cm^3), $\sigma(E)$ is the ^{10}B neutron capture cross section (in cm^2) for the energy E, ε is the energy of the corresponding capture reaction (2.31 MeV) and k is a unit conversion constant, from MeV to J.

2.3. Measurements with activation foils

The thermal ($E < 0.1$ eV) and epithermal (0.1 eV $< E <$ 100 keV) neutron flux in three irradiation channels of the nuclear reactor were measured with Gold and Manganese-Nickel foils. Some of the gold foils were placed inside cadmium capsules, so they were activated only with epithermal neutrons. Gamma activity in gold and manganese foils were measured with a sodium iodide scintillator detector coupled with a multi-channel analyzer. The flux for neutrons with energies $E < 0.1$ eV and with 0.1 eV $< E <$ 100 keV were determined following the standard Westcott formalism (Westcott, 1955).

The overall uncertainty for activation foils was determined

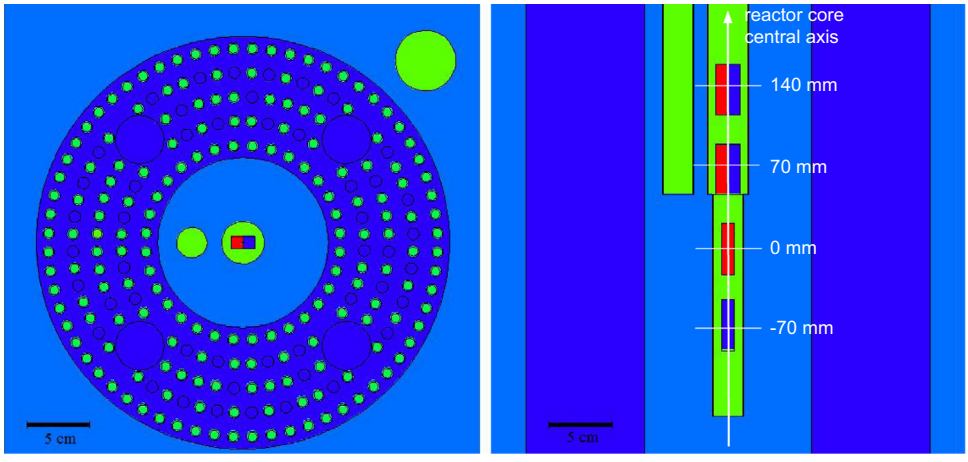

Fig. 1. Top (left) and lateral (right) view of the nuclear reactor RA-0 core. The ^{10}B-doped cuvettes are represented in blue and the standard samples in red. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

similar to Fricke gel dosimeters:

$$(\sigma_{activation\ foils})^2 = (\sigma_{position})^2 + (\sigma_{gamma\ activity})^2$$

where *position* refers to foils positioning inside the irradiation channel in the nuclear reactor and *gamma activity* refers to the determination of the neutron flux from the activity of the irradiated foils.

3. Results and discussion

3.1. MCNP results

Simulations using the main code MCNP were carried out in order to verify that the addition of Fricke gel dosimeters does not significantly alter neutron transport in the nuclear reactor core. Different simulations were run with and without dosimeters inserted in the reactor core. For these simulations, standard and Fricke gel dosimeter with 400 ppm of ^{10}B cuvettes were used. The thermal flux ϕ_{th} and epithermal flux ϕ_{epi} were calculated. The obtained results are depicted in Fig. 2.

As can be appreciated in Fig. 2, the differences are below 5% in the regions where the dosimeters were placed (see also Fig. 1), thus indicating minimal perturbation of radiation field. These differences are due to the increase of absorption of thermal neutron in the boron added to the dosimeters located at −70 mm and those located at 70 and 140 mm.

Similarly, neutron dose estimation and neutron flux/spectrum were simulated for each cuvette location. Results for central (0 mm) and top (140 mm) locations are reported in Fig. 3. Differences in neutron flux due to the presence of boron are found to be less than 10%.

In order to study the contributions of gamma-rays and neutrons to the total dose, simulations were performed changing the concentration of ^{10}B in the Fricke material. According to the characteristics of the RA-0 reactor, it was decided that 0, 200, 400, 600 and 800 ppm of ^{10}B would be used to study the effect of the absorbed dose component due to thermal neutrons. The results obtained by MC simulation for dose components due only to the presence of ^{10}B (D_{th}), total neutron dose and photon dose are shown in Fig. 4.

The corresponding uncertainties for these calculations are, on average, 3%. As can be seen, the photon dose remains constant, independent from the ^{10}B concentration. Total neutron dose increase is given by the sum of the total neutron dose at 0 ppm of ^{10}B and the boron dose for each concentration. Also, for 400 ppm of ^{10}B the dose due to boron capture is higher than the gamma dose. Therefore, this concentration was chosen for Fricke gel dosimeter elaboration.

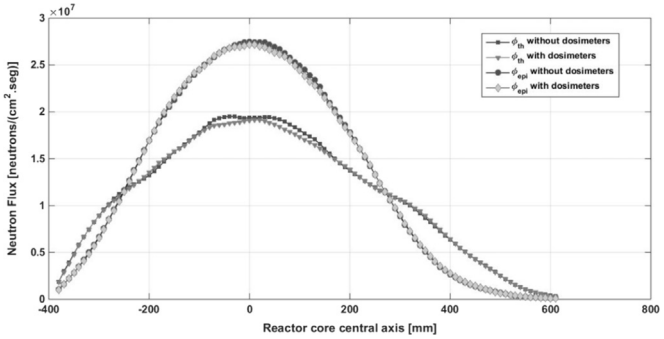

Fig. 2. Simulated neutron flux in the reactor central axis. Data uncertainty is in average under 3%.

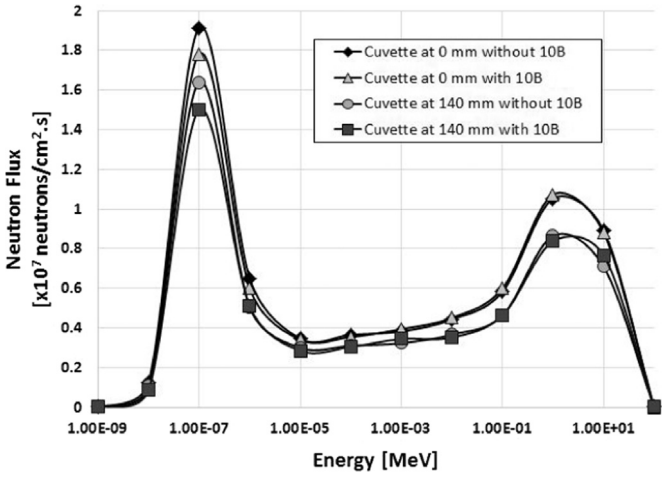

Fig. 3. Simulated neutron flux for each cuvette location. Points uncertainties are, in average, less than 3%.

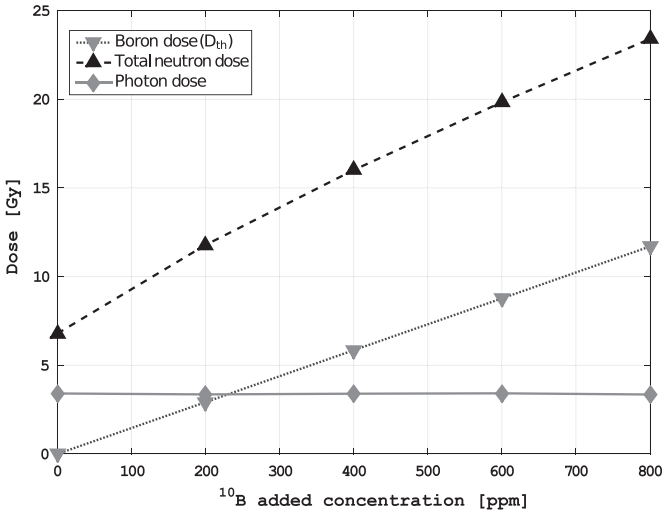

Fig. 4. Contributions to the absorbed dose obtained by simulations in a cuvette volume as function of ^{10}B concentration.

3.2. Characterization of Fricke gel dosimeters

In order to verify that the addition of boron does not change the optical absorption properties of the Fricke gel dosimeters, the absorbance spectrum was analyzed. Some illustrative results are shown in Fig. 5. The measurements were made before and after irradiation with X-rays, and also before and after irradiation at the nuclear reactor.

As can be seen in Fig. 5, the wavelength of maximum absorption is not altered by the boron presence, but the relative absorption between borated and standard (non-borated) samples is appreciable. This fact does not represent a drawback because separate calibrations curves are needed, as depicted in Fig. 6.

The calibration of the standard and borated dosimeters irradiated with X-ray beams was made integrating the optical absorbance spectrum around its maximum value for samples irradiated from 0 Gy to 30 Gy. Good linear correlation was obtained in both linear fits: $D_{0\ ppm\ ^{10}B} \approx (62.5 \pm 0.6)\ \Delta OD_{0\ ppm\ ^{10}B}$ and $D_{400\ ppm\ ^{10}B} \approx (18.6 \pm 0.9)\ \Delta OD_{400\ ppm\ ^{10}B}$ and the linear correlation factors are 0.99 and 0.98 for 0 and 400 ppm, respectively.

3.3. Measurements in the nuclear reactor

For the irradiation at the RA-0, Fricke dosimeters were placed

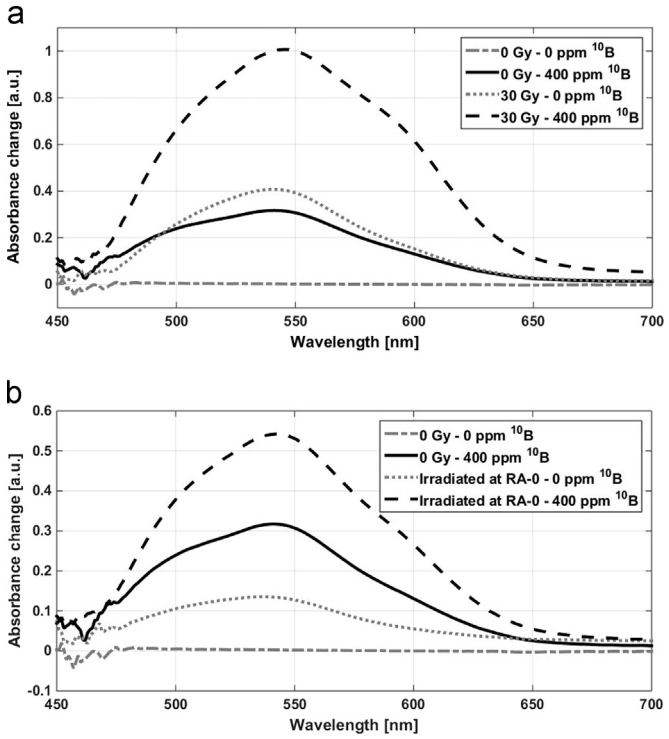

Fig. 5. Light absorption spectra of Fricke gel dosimeters irradiated with X-ray beams (a - top) and in nuclear reactor mixed field (b - bottom).

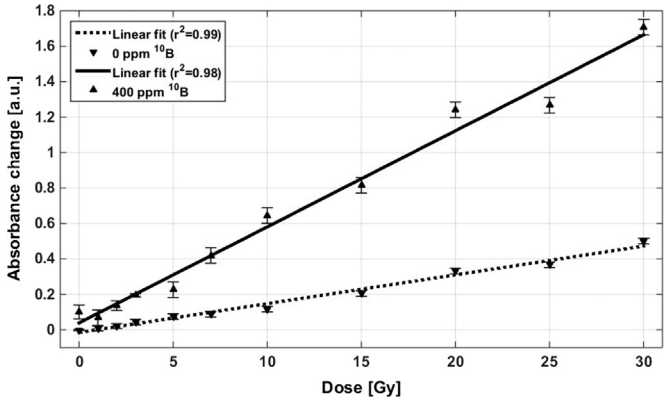

Fig. 6. Calibration curves for borated and standard Fricke gel dosimeters irradiated with X-rays.

in the locations indicated in Fig. 1. By means of Eq. (1) and with the relative sensitivity factors, from Fricke gel dosimeters measurements the dose rate at the reactor core was calculated. Similarly, neutron fluxes measured by activation foils were used to estimate the corresponding dose rate using expression (2). Fig. 7 reports the final results regarding the dose rate evaluated in different positions along the central axis of the RA-0 core.

Fricke gel measurements (circles) are in agreement, within experimental uncertainties, with theoretical calculation from activation foils measurements (triangles) and MCNP results (squares). Actually, it is remarkable that dose rate estimations by Fricke gel dosimeters, activation foils and MC calculations are statistically indistinguishable. Nevertheless, improvements are needed in the gel dosimetry technique in order to reduce measurement uncertainties.

4. Conclusions

Monte Carlo simulations using MCNP code were carried out in order to obtain different neutron and gamma information about the RA-0 zero power nuclear reactor. Fricke gel dosimeter measurements, including ^{10}B, were carried out. Boron neutron capture contribution to total absorbed dose could be separated in both methods. Results for both MCNP simulations and ^{10}B doped Fricke gel dosimetry are in good agreement also with activation foil measurements.

From the results reported in this work, it can be concluded that Fricke gel dosimeters have a promising potential for the characterization of thermal neutron dose component inside the core of a zero power nuclear reactor.

It was possible to obtain reliable measurements and calculations about the dose rate inside the core of the zero power RA-0 nuclear reactor. Further work is needed in order to continue improving the technique of Fricke gel dosimetry while reducing uncertainties of neutron dose measurements.

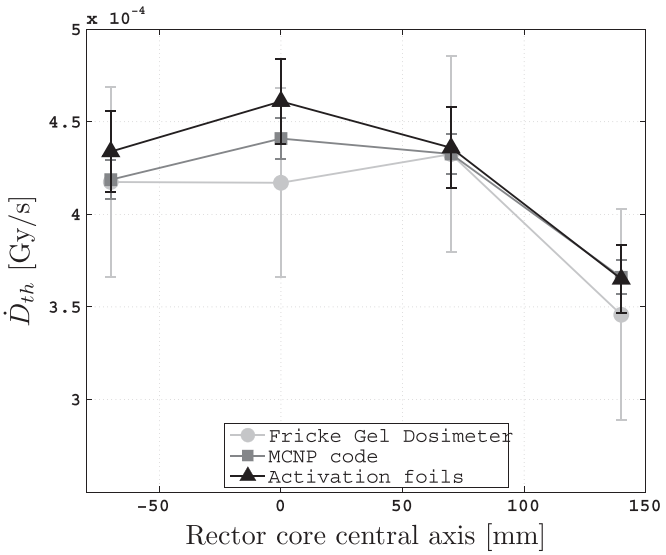

Fig. 7. Neutron dose rate due to neutron capture by ^{10}B ($\dot{D}_{th}$) inside the RA-0 core, measured with Fricke Gel Dosimeter (circles), obtained from activation foils (triangles) and calculated with MCNP code simulations (squares). Uncertainties correspond to one standard deviation.

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