

Use of the chemical Fricke dosimeter and its modifications for dosimetry of gamma neutron radiation of a pulsed reactor*

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Abstract

The paper investigates the characteristics of the chemical Fricke dosimeter (with the standard composition (D1), without NaCl addition to the solution (D2), without NaCl but with a tenfold increased concentration of Fe²⁺ (D3)) under continuous and pulsed irradiation with an ultra-high dose rate of the BARS-6 reactor with unshielded metallic cores.

The dosimeter radiosensitivity had a linear dependence on the gamma neutron radiation dose in a range of 25 to 750 Gy and was respectively $1.96 \pm 0.05 \mu\text{Gy}^{-1}$ (D1), $2.04 \pm 0.05 \mu\text{Gy}^{-1}$ (D2), and $2.08 \pm 0.5 \mu\text{Gy}^{-1}$ (D3) in the continuous irradiation mode, and $1.24 \pm 0.05 \mu\text{Gy}^{-1}$, $2.00 \pm 0.05 \mu\text{Gy}^{-1}$, and $1.94 \pm 0.05 \mu\text{Gy}^{-1}$ in the pulsed irradiation mode. This makes $\approx 60\%$ of their sensitivity to the ⁶⁰Co gamma radiation ($3.40 \pm 0.02 \mu\text{Gy}^{-1}$), and 36%, 1.6 times as less, for a standard Fricke dosimeter irradiated in the pulsed mode. The experimental value of the radiation chemical yield, $G_n(\text{Fe}^{3+})$, for all solution modifications and both irradiation modes varied slightly and was $0.84 \pm 0.11 \mu\text{M/J}$ on the average, except for the standard solution in the pulsed mode ($0.66 \pm 0.07 \mu\text{M/J}$). The neutron doses determined by chemical and activation dosimeters coincided within the error limits, but the chemical dosimeter readings were systematically higher, by about 20%.

Therefore, in the fission spectrum neutron dose rate range of 0.4 to 7×10^8 Gy/min, there is no dose rate effect both in the standard Fricke dosimeter version (without NaCl) and in the modified version, which makes it possible to use modified Fricke dosimeters to assess the physical and dosimetry characteristics of mixed gamma neutron radiation beams.

Keywords

Ferrous sulfate Fricke dosimeter, fast neutrons, gamma radiation, ultrahigh dose rate, BARS-6 pulsed reactor

Introduction

At the present time, practically all types of electromagnetic and ionizing radiation are used in medicine, specifically

in oncology. Units with radiation of high linear energy transfer (LET), thanks to their higher relative biological effectiveness (RBE) as compared with standard gamma radiation, have been used on an increasingly growing

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scale in radiation therapy of patients with radioresistant malignant tumors of different localization. Primarily, these are accelerators of carbon-12 ions (Kaprin and Ulyanenko 2016). A niche of their own, not large for the time being, is occupied by neutron radiation sources the efficiency of which in combined gamma neutron irradiation regimens has been shown in a number of studies (Vazhenin et al. 2007, Bobkova et al. 2012, Kandakova 2015, Musabaeva et al. 2016), including in treatment of oncologic patients at the BR-10 reactor by doctors of the Tsyb Medical Radiological Research Center (Tsyb et al. 2003, Gulidov et al. 2004, Gulidov and Mardynskiy 2006).

There is a growing interest in investigating pulse neutron impacts explained both by fundamental aspects and by certain practical tasks of current concern. In fundamental terms, this is exploration of fast biological and radiobiological processes and determination of the RBE for radiations with a high and ultrahigh dose rate (Vazhenin et al. 2007, Koryakina 2014). Application tasks are highly diverse. The practical use of radiations with an ultrahigh dose rate, including pulse radiations, to increase the efficiency of beam therapy, the so-called flash therapy, is considered theoretically (Symonds and Jones 2019, Jin et al. 2020, Marlen et al. 2020). This leads to new dosimetry tasks which are addressed, along with ionization techniques, with the extensive use of methods based on using liquid and solid scintillators, semi-conductors, and luminescent and chemical dosimeters. The latter use chemical radiation-induced changes taking place in certain substances. One of such dosimeters is the Fricke chemical dosimeter used in gamma radiation and electron dosimetry in a dose range of 0.05 to 2000 Gy and a dose rate range of up to 250 Gy/min (Sokolova 1972, Pikaev 1975).

Fast and intermediate neutron beams are often accompanied by gamma quanta, so the major problem in dosimetry of fast and intermediate neutrons consists in separating the effect from the associated gamma radiation since their radiobiological effect is different. This, in most cases, is done using two types of dosimeters one of which has the smallest possible sensitivity to neutrons and allows estimating the contribution of gamma radiation. In the case of fast neutron action on a biological object, the absorbed dose is due predominantly to recoil protons, so aqueous solutions of any compounds or organic compounds close to tissue in terms of composition are a choice for use as chemical systems. A ferrous sulfate system, the most widespread one among aqueous solutions of inorganic compounds, meets this requirement but exhibits a dependence on radiation LET so it is used for precision measurements of the fast neutron dose when the energy spectrum of fast neutrons is known. The dependence of radiation chemical yield, $G(\text{Fe}^{3+})$, on neutron energy in a range of 0.1 to 14 MeV is presented in (Sokolova 1972). It is shown that it varies in the limits of $\pm 6\%$ in a range of 0.1 to 1.5 MeV, and in the limits of $\pm 15\%$ in a range of 1.5 to 14 MeV.

Where the neutron spectrum is not known, a ferrous sulfate dosimeter makes it possible to measure tissue

doses of intermediate and fast neutrons ($E_n < 20$ MeV) with an additional error of $\pm 20\%$ and with a smaller error if the available spectrum data is limited (Sokolova 1972). In conjunction with any other method with low sensitivity to neutrons as compared with sensitivity to gamma radiation, e.g., thermoluminescent method, this system allows determining the total tissue dose of gamma neutron radiation and its neutron contribution.

The purpose of the study was to investigate the performance of chemical Fricke dosimeters with different compositions when irradiated by continuous and pulse BARS-6 reactor radiation with an ultrahigh dose rate.

Materials and methods

Preparation of chemical dosimeter solutions

To prepare a standard dosimeter Fricke solution, 550 mg of Mohr's salt was dissolved in a small amount of triple-distilled water, with an addition of 55 mg NaCl, and mixed until dissolved in full. Sulfuric acid (22 ml) was added to the resultant solution with the dosimeter solution volume brought to 1 l using triple-distilled water. A standard solution without NaCl addition was also prepared for the BARS-6 pulsed reactor irradiation, since its presence can reduce the Fe^{3+} ion yield, $G(\text{Fe}^{3+})$ (Pikaev et al. 1963, Pikaev 1975, Klassen et al. 1999), and a modified Fricke dosimeter was used for the dosimetry of radiation with an ultrahigh dose rate ($\sim 1 \times 10^6$ Gy/min), in which, apart from excluding NaCl from the composition, the Fe^{2+} iron concentration was increased tenfold, to 1×10^{-2} M. The prepared solutions were left for 12 h in a dark place at room temperature and then placed in a refrigerator (the dosimeter readings are stable for one year during refrigerated storage). Prior to being used, the dosimeter solutions were let to be heated to room temperature.

Calibration of dosimeter solutions

The dosimeter parameters of the prepared Fricke solutions were tested for being compliant with the standard characteristics under ^{60}Co gamma radiation (the dose rate is ~ 43 Gy/min) in a dose range of 10 to 150 Gy. The optical density of the irradiated solutions was measured at the 304 nm wavelength with the slit's spectral width of 2.5 nm in quartz cuvettes with the absorbing layer thickness of 1 cm. A non-irradiated dosimeter system was used as the test solution for the measurements.

Neutron irradiation

The Fricke dosimeters were irradiated in plastic test tubes of 4 ml by unfiltered radiation in a pulsed mode and in a continuous mode of the BARS-6 reactor operation (IPPE, Obninsk). The test tubes with the dosimeter solutions (Fig. 1) were attached to a vertical rod installed between the cores, at a height of 100 to 1500 mm above the axis

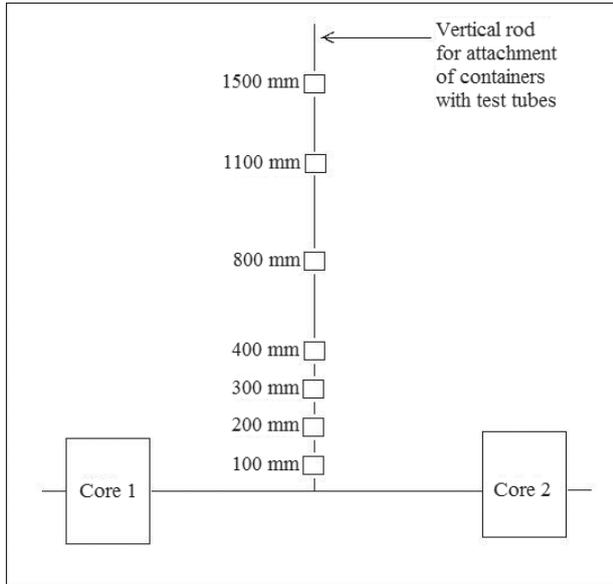


Figure 1. Irradiation geometry of chemical Fricke dosimeters at the BARS-6 reactor in pulsed and continuous modes.

connecting the core centers. They were placed at the same positions in both irradiation modes which ensured that the physical dosimetry conditions of irradiation were identical.

The irradiation time in the continuous mode of the reactor operation was ~ 60 min, and that in the single pulse mode was ~ 65 to 70 μ s. At the Fricke dosimeter positions, depending on the distance from the reactor cores, the neutron doses changed in a range of ~ 25 to 750 Gy with the dose rate being 0.4 to 13 Gy/min in the continuous mode and $(0.2 \text{ to } 7) \cdot 10^8$ Gy/min in the pulse mode. The number of divisions in the cores was approximately equal for both modes in paired experiments and was monitored by the reactor personnel with an accuracy of $\pm 2\%$ (Koryakina 2014). The absorbed tissue neutron and gamma radiation doses were measured by the reactor personnel using nickel activation detectors and thermoluminescent dosimeters. The measurement error was estimated to be 16% (D_n) and 30% (D_γ) at the 95% confidence level (Prokhorov et al. 1998).

Calculation of doses and radiation chemical yield for a dosimeter solution in the mixed gamma radiation field

For the dosimetry of mixed gamma neutron radiation by a standard method with a pair of dosimeters (PikaeV 1975), chemical dosimeters represent a component of a pair which is more sensitive to neutrons. An IKS-A thermoluminescent dosimeter (TLD) was used as the dosimeter sensitive to gamma radiation (Bochvar et al. 1972). Taking into account that the chemical dosimeter readings (optical density) represent a sum of the neutron component and the gamma component, we have a system of two equations for determining the neutron and gamma components of the mixed radiation dose:

$$S_{\gamma+n}^{\text{P-P}} = S_{\gamma}^{\text{P-P}} + S_n^{\text{P-P}} = \alpha_{\gamma}^{\text{P-P}} D_{\gamma} + \alpha_n^{\text{P-P}} D_n, \quad (1)$$

$$D_{\text{TLD}} = \alpha_{\gamma}^{\text{TLD}} D_{\gamma} + \alpha_n^{\text{TLD}} D_n, \quad (2)$$

where $S_{\gamma+n}^{\text{P-P}}$ is the relative optical density of the dosimeter solution; $S_{\gamma}^{\text{P-P}}$ and $S_n^{\text{P-P}}$ are the optical densities due to the gamma- and neutron component of the dose, respectively; $\alpha_{\gamma}^{\text{P-P}}$, $\alpha_n^{\text{P-P}}$ are the chemical dosimeter and TLD sensitivity to gamma radiation; $\alpha_{\gamma}^{\text{TLD}}$, α_n^{TLD} are the chemical dosimeter and TLD sensitivity to neutrons; D_{TLD} is the dose according to the TLD readings; and D_{γ} , D_n are the doses of the gamma and neutron components of the total dose.

Solving system of equations (1), (2), we get

$$D_n = (S_{\gamma+n}^{\text{P-P}}/\alpha_{\gamma}^{\text{P-P}} - D_{\text{TLD}}) / (G_n/G_{\gamma} - \alpha_n^{\text{TLD}}/\alpha_{\gamma}^{\text{TLD}}), \quad (3)$$

where G_n , G_{γ} are the radiation chemical yield of Fe^{3+} ions after exposure to neutron and gamma radiation.

Sensitivity of the chemical dosimeter to gamma radiation and neutrons can be written as

$$\alpha_{\gamma} = \Delta S_{\gamma} / D_{\gamma} = G_{\gamma}(\text{Fe}^{3+})\epsilon/\rho, \quad (4)$$

$$\alpha_n = \Delta S_n / D_n = G_n(\text{Fe}^{3+})\epsilon/\rho = \alpha_{\gamma} G_n(\text{Fe}^{3+})/G_{\gamma}(\text{Fe}^{3+}). \quad (5)$$

The Fricke dosimeter sensitivity to gamma radiation is $34.9 \times 10^{-4} \text{ Gy}^{-1}$ with standard parameters: $G(\text{Fe}^{3+}) = 1.607 \times 10^{-6} \text{ mol/J}$; molar coefficient of Fe^{3+} ion extinction at a wavelength of 304 nm and a temperature of 20 °C $\epsilon = 2121 \text{ l}/(\text{mol} \times \text{cm})$; absorbing layer thickness $l = 1 \text{ cm}$; solution density $\rho = 1.024 \text{ kg/l}$.

$$G_n(\text{Fe}^{3+}) = \Delta S_n / (\epsilon/\rho D_n). \quad (6)$$

To calculate neutron doses using formula (3) we determine the average G_n value on the base of the energy dependence, $G_n(E)$ (Lawson and Porter 1975, Kapchigashev et al. 1984) for the neutron spectra from the BARS-6 reactor cores at irradiation points using own calculations and published data (Kurachenko et al. 2008):

$$G_n(E) = \sum_E G_n(E) \cdot K(E) \cdot N(E) dE / \sum_E K(E) \cdot N(E) dE, \quad (7)$$

where $K(E)$ is the kerma in the ferrous sulfate solution for the neutrons with energy E ; and $N(E)dE$ is the number of neutrons with energies in the range from E to $E+dE$.

The shapes and dimensions of the reactor cores, their positions in the reactor hall and its dimensions and walls (Prokhorov et al. 1998, Kurachenko et al. 2008), and the arrangement geometry of the irradiated samples were taken into account in the neutron spectra calculation (using the MCNP-5 code). The relative group error for the neutron spectrum calculation did not exceed 5% for the range from 0.1 to 4 MeV and 10% for the range from 4 to 10 MeV. The statistical error of the energy total fluence is $\leq 1.5\%$.

The allowance for the temperature dependence of the dosimeter solution radiosensitivity during the irradiation and in the process of the optical density measurement which is directly proportional to that of the molar extinction coefficient was made based on data in (PikaeV 1975).

Statistical analysis

Standard statistical analysis methods were used for the experimental data processing. An analysis of the dose dependences was undertaken using a linear regression model by the least squares method (Microcal Origin 6.1). The error of the variables of type $Z = f(x_1, \dots, x_k)$, where $Z = D_n(\text{Fe})$ or $G_n(\text{Fe}^{3+})$, was estimated using the partial derivatives method for the case of the error's uncorrelated components:

$$\sigma(Z) = \sqrt{\sum_i^k \sigma_i^2(Z)} = \sqrt{\sum_i^k \left(\frac{\partial Z}{\partial x_i}\right)^2 \sigma^2(x_i)}. \quad (8)$$

The statistical significance of results and the differences between them were estimated using Student's *t*-criterion.

Results and discussion

Prior to using dosimeter solutions to investigate their characteristics in the reactor irradiation pulse fields, it was necessary to make sure that their dosimeter parameters following exposure to gamma radiation complied with standard ones. To this end, the chemical dosimeters used in the study were calibrated at an irradiation unit with a ^{60}Co source with known dosimeter characteristics.

Fig. 2 presents dose dependences of the optical density for the three Fricke dosimeter versions. In the investigated range of doses, the optical densities of the dosimeters are approximated by straight lines through the origin of coordinates which coincide within the error limits. The coefficients of linear regression equations are $(34.1 \pm 0.2) \times 10^{-4} \text{ Gy}^{-1}$ for the standard solution, $(33.9 \pm 0.7) \times 10^{-4} \text{ Gy}^{-1}$ for the solution without NaCl in the composition, and $(33.6 \pm 0.5) \times 10^{-4} \text{ Gy}^{-1}$ for the solution with the Fe^{2+} ion concen-

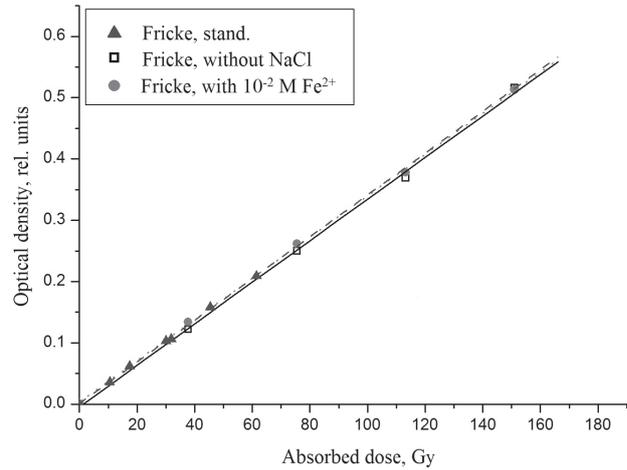


Figure 2. Optical density for three Fricke dosimeter versions as a function of the ^{60}Co gamma radiation dose.

tration of $1 \times 10^{-2} \text{ M}$ and without NaCl. With an accuracy of up to 3.5%, the experimental data coincide with the value of the standard Fricke dosimeter solution sensitivity to the ^{60}Co radiation ($34.9 \times 10^{-4} \text{ Gy}^{-1}$) (PikaeV 1975). The differences between the experimental values and the values taken from literature can be caused by a number of factors: the ^{60}Co radiation dose measurement error (10% at $P = 0.95$), difference in the $\epsilon(\text{Fe}^{3+})$ value for the spectrophotometer used in the study from the standard one; trace impurities in chemical reagents, and their combination.

The results of the experiments to study the effects of the irradiation mode (pulsed or continuous) on the standard chemical Fricke dosimeter readings and solutions with a modified composition are shown in Fig. 3.

The regression dependence of the dosimeter solution optical density on the reactor's integral absorbed radiation dose is linear for all irradiation options. And the optical density of the standard Fricke dosimeter with doses below

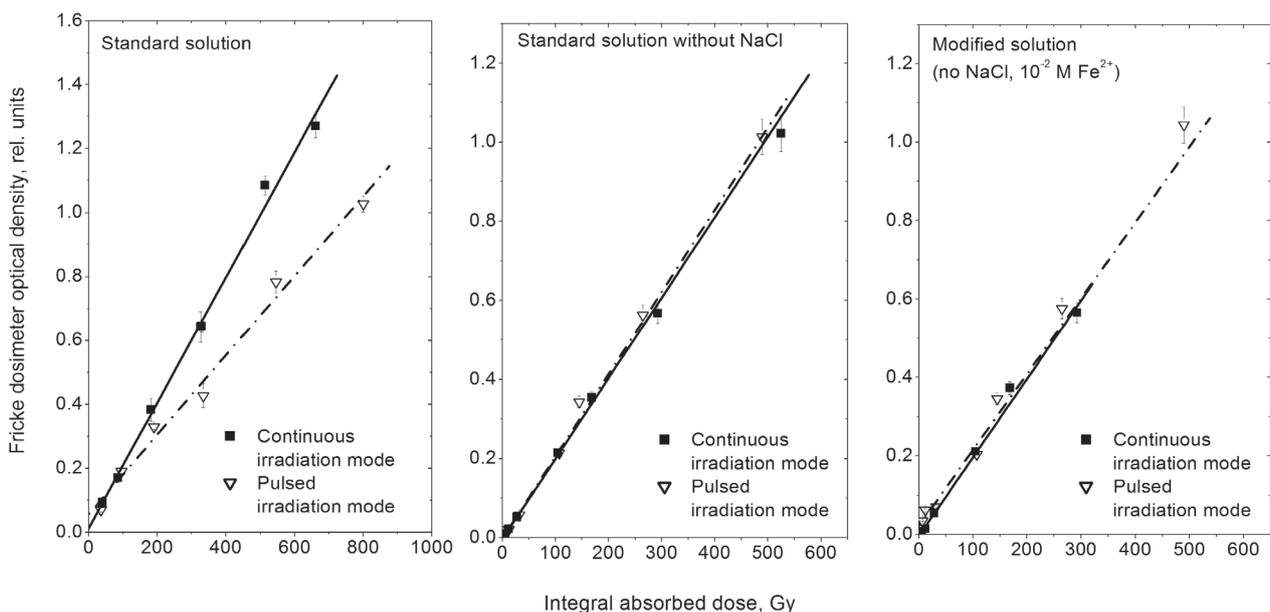


Figure 3. Optical densities for three Fricke dosimeter versions as a function of neutron dose during BARS-6 irradiation in continuous and pulsed modes.

100 Gy coincides with both irradiation modes. With higher doses, the optical density in the pulsed irradiation mode decreases by a factor of 1.5 (the curve slopes are respectively $(19.6 \pm 0.5) \times 10^{-4} \text{ Gy}^{-1}$ and $(12.4 \pm 0.5) \times 10^{-4} \text{ Gy}^{-1}$). The readings of the dosimeter solutions without NaCl, as well as with an increased concentration of Fe^{2+} do not practically differ one from another both during continuous irradiation ($(20.4 \pm 0.5) \times 10^{-4} \text{ Gy}^{-1}$ and $(20.0 \pm 0.5) \times 10^{-4} \text{ Gy}^{-1}$) and during pulsed irradiation ($(20.8 \pm 0.5) \times 10^{-4} \text{ Gy}^{-1}$ and $(19.4 \pm 0.5) \times 10^{-4} \text{ Gy}^{-1}$), as well as from the readings of a standard Fricke dosimeter in the continuous irradiation mode.

The sensitivity of the dosimeter solutions to reactor radiation was about 60% during irradiation in the continuous mode (and in the pulsed mode for solutions with a modified composition), and about 40% for the standard solution in the pulsed mode. The major component of reactor radiation in the experiment conditions was neutron and the contribution of gamma radiation to the total dose was in a range of $\approx 5\%$ at distances of ≤ 0.3 m from the cores and up to 18% and more at distances of > 0.85 m due to a relative increase of the secondary gamma radiation from the walls to the total dose. The results obtained confirm that a Fricke dosimeters can be used for dosimetry of high-intensity fluxes of neutron and mixed gamma neutron radiation with a slight modification of the solution composition (no NaCl in the solution and an additional increase in the concentration of Fe^{2+}) (Pikaev et al. 1963, Pikaev 1975). The use of solutions without NaCl, however, increases the requirements to the quality of chemical reagents, the solution preparation technology and the experiment conditions so that to prevent organic impurities from accidentally entering the solution.

Important in using a Fricke dosimeter for dosimetry of mixed gamma neutron radiation is to identify the contribution of gamma radiation to the total dose and the radiation chemical yield value of Fe^{3+} for the dose's neutron component. Thanks to the fact that there were witness dosimeters (nickel detectors and IKS TLDs (Bochvar et al. 1972, Prokhorov et al. 1998)) placed at each irradiation point of the test tubes with the dosimeter solution, the neutron tissue kerma and gamma radiation tissue dose values were calculated based on their readings. Experimental values $G_n(\text{Fe}^{3+})$ and relation G_n/G_γ were determined for different Fricke dosimeter composition versions based on formulas (1) and (7). During the continuous mode irradiation, the value of G_n/G_γ was respectively 0.54 (100%), 0.52 (3%) and 0.51 (6%) for the Fricke dosimeter with the standard composition without NaCl and with an increased concentration of ferrous iron and also without NaCl. During the pulsed mode irradiation, the respective values of G_n/G_γ were 0.41 (25%), 0.51 (5%) and 0.54 (0%). As can be seen, the value of $G_n(\text{Fe}^{3+})$ for all solution composition versions and both irradiation modes changed slightly (excluding data for the standard solution in the pulsed mode) and was $0.84 \pm 0.11 \mu\text{M/J}$ on the average. The estimated error of the radiation chemical yield determination is 11% ($P = 0.63$) and is due primarily to the statistical error of the neutron dose, as well as that of the partial optical density of the dosimeter solution following exposure to neutrons. The latter are caused by the fast neutron fluence

determination errors (8% (Prokhorov et al. 1998)), by the error of the specific fission spectrum neutron tissue kerma value (5%), the gamma radiation dose (15% (Prokhorov et al. 1998)), the spectrophotometer ($\leq 1\%$), and the molar extinction coefficient (2%). With regard for the error, the values of the radiation chemical yield, $G_n(\text{Fe}^{3+})$, in solutions with different compositions during irradiation in both modes do not differ, including for the solution with the standard composition, the optical density of which differed by a factor of 1.5 during the continuous and pulsed mode irradiations.

For a fission neutron spectrum of ^{252}Cf ($E_{\text{av}} = 2.15 \text{ MeV}$), value $G_n(\text{Fe}^{3+}) = 0.78 \pm 0.11 \mu\text{M/J}$ or $G_n/G_\gamma = 0.48$ was determined (Greene et al. 1973). This is 10.5 to 12% as low as the values obtained in the study for the BARS-6 reactor neutrons, the average leakage spectrum energy of which is $\leq 1.44 \text{ MeV}$ (Prokhorov et al. 1998, Kurachenko et al. 2008) but fits these with regard for the mutual errors of 22 and 14.7%. For a spectrum of the BR-10 reactor B-3 beam fission neutrons with a less average energy of 0.85 MeV, experimental value $G_n(\text{Fe}^{3+}) = 0.70 \pm 0.04 \mu\text{M/J}$ or $G_n/G_\gamma = 0.44$ (Kapchigashev et al. 1984). The $G_n(\text{Fe}^{3+})$ values, overestimated as compared with data in (Greene et al. 1973, Kapchigashev et al. 1984), can be explained, as it follows from formula (7), by the experimental underestimates of the gamma radiation and neutron doses. The calculation results (Kurachenko et al. 2008) show that gamma quanta with an energy of 1 to 10 MeV constitute up to a half of the total spectrum which may lead to the dose being underestimated by the IKS-A TLD due to the absence of electronic equilibrium conditions. Using a threshold nickel detector ($E_{\text{thr}} = 2.5\text{--}3 \text{ MeV}$) to estimate the fluence of the total fission neutron spectrum (Prokhorov et al. 1998) and the kerma for the total spectrum could also lead to the neutron dose underestimation which was observed, e.g., at large distances from the cores (in excess of 6 m) (Koryakina 2014).

The value of the radiation chemical yield, $G_n(\text{Fe}^{3+})$, for neutrons was calculated also based on its dependence on the neutron energy (Lawson and Porter 1975, Kapchigashev et al. 1984) for the neutron spectra at the dosimeter solution locations. At distances from 0.25 to 0.6 m from the reactor cores, the spectra changed slightly and the calculated value of $G_n(\text{Fe}^{3+})$ changed by not more than 1.5%. The average value of $G_n(\text{Fe}^{3+})$ for the solutions irradiated at these distances was 0.76 and 0.78 $\mu\text{M/J}$ based on data in (Lawson and Porter 1975, Kapchigashev et al. 1984), respectively, or $G_n/G_\gamma = 0.475$ and 0.485. According to estimates (Lawson and Porter 1975), the total error of value $G_n(\text{Fe}^{3+})$ was about 3% above 3.5 MeV, 8% at energies of about 1 MeV and 30% at energies 0–1 MeV. For the fission neutron spectrum, the error is estimated to be 10 to 12% ($P = 0.95$) (Lawson and Porter 1975).

The calculated values of $G_n(\text{Fe}^{3+})$ and G_n/G_γ make it possible to determine the BARS reactor neutron dose using formula (4) and the IKS-A TLD measurement results. Thus obtained neutron doses were compared with the doses determined using nickel detectors (Prokhorov et al. 1998, Koryakina 2014). The results are presented in Fig. 4 for two irradiation modes (pulsed and continuous) for a standard Fricke dosimeter, a standard Fricke dosimeter with no NaCl

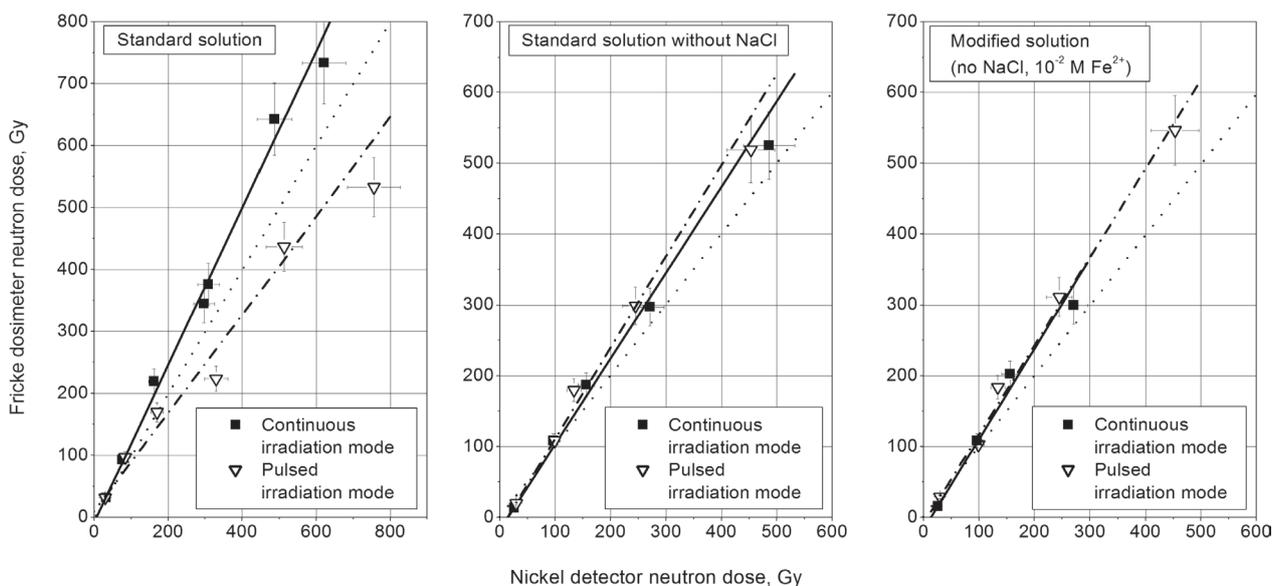


Figure 4. Relation between neutron doses obtained based on readings of the chemical dosimeters (the standard dosimeter, the standard dosimeter without NaCl addition, and with an increased concentration of Fe^{2+} ions) and the nickel detector during continuous and pulsed mode irradiation. The dashed line corresponds to the equality of doses determined by two methods.

in the composition and that with an increased Fe^{2+} iron concentration. The estimated calculation error is 20% ($P = 0.95$).

During the irradiation of the standard solution in the continuous mode and of the standard solution without NaCl and the modified solution in both modes, the values of the neutron doses determined by the chemical dosimeter systematically exceed the dose value calculated based on the activation detector measurements. The slopes of the regression curves in the continuous mode are 1.27 ± 0.06 (standard solution), 1.21 ± 0.06 (standard solution without NaCl) and 1.27 ± 0.07 (modified solution) and those in the pulsed mode are 0.76 ± 0.04 , 1.29 ± 0.07 and 1.25 ± 0.07 , respectively. With regard for the errors, the differences of relation $D_{\text{Fe}}/D_{\text{Ni}}$ from 1.0, according to Student's criterion, are statistically significant for the standard Fricke dosimeter for both irradiation modes ($P > 0.99$), but the calculated dose values for the standard solution without NaCl and the modified solution did not differ noticeably from those determined by activation method. Using a Fricke dosimeter with the standard composition (with 1×10^{-3} M NaCl) to measure the fission spectrum neutron dose with a high dose rate ($> 1.5 \times 10^8$ Gy/min) leads to a neutron dose underestimation by a factor of 1.6, on the average, as compared with irradiation with a lower dose rate. Approximately the same threshold dose rate value, at which $G(\text{Fe}^{3+})$ starts to decrease, was observed following electron irradiation (about 1×10^8 Gy/min (Pikaev 1975)). Therefore, a careful approach is required to use a standard chemical Fricke dosimeter for dosimetry of pulsed neutron radiation with a dose rate in excess of 1×10^8 Gy/min. At the same time, excluding NaCl from the solution composition and increasing the concentration of Fe^{2+} to 1×10^{-2} M leads to the ferrous sulfate dosimeter system readings being independent in the neutron dose rate range from 0.2×10^8 to 7×10^8 Gy/min and the dose range of up to 600 Gy (Fig. 4), which confirms that recommendations

with respect to using such system for dosimetry of pulsed irradiation (Pikaev 1975, Klassen et al. 1999).

The noticeable discrepancies in the BARS-6 reactor neutron dose estimates in the limits of 20 to 30%, based on activation and chemical dosimetry methods, with the total error for each of the methods being about 20% ($P = 0.95$), make it difficult to obtain reliable estimates of the radiobiological parameters for biological objects (e.g., the relative biological efficiency of pulsed radiation) in experiments using this unique source of gamma neutron radiation. An approach has turned out to be fruitful which involves paired experiments with irradiation in a pulsed mode and a continuous mode with an interval of one day when irradiated biological or other samples are placed at the same positions which are reproduced with rather a high degree of accuracy ($< 0.5\%$). And the number of fissions in the reactor cores is selected to be approximately equal for both irradiation modes and is controlled by personnel with an accuracy of not worse than 2%. This does not, however, exclude the need for improving the accuracy of activation and chemical dosimeters, including through a more extensive use of Monte Carlo simulations both to calculate the neutron and gamma radiation spectra and energy deposition (Kurachenko et al. 2008) and the radiolysis of water and aqueous solutions, including a Fricke dosimeter, and its dependence on LET and the type of charged particles (Autsavapromporn et al. 2007). With a theoretical estimate of $G_{\text{n}}(\text{Fe}^{3+})$, it is also important to know the effective spectra of secondary charged particles at the chemical dosimeter irradiation point (Bochvar et al. 1972) by calculating these for particular conditions of the dosimeter solution irradiation, and the energy dependence of $G_{\text{n}}(\text{Fe}^{3+})$ for particles with $Z = 1-6$ at an energy of less than 20 MeV and, especially, below 0.2 MeV where experimental data are absent.

Conclusion

The results of the studies have shown that a chemical Fricke dosimeter system and its modifications (without NaCl in the

composition both with the standard concentration and the concentration of Fe^{2+} iron increased to 1×10^{-2} M) can be used for dosimetry of mixed gamma neutron radiation of reactors (including a pulsed reactor) in a broad dose rate range of 0.4 to 7×10^8 Gy/min and a dose range of up to 750 Gy.

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