

Study of the X-Ray Radiation in Metal-Containing Composites Irradiated with Neutrons and Photons

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Abstract—Secondary X-ray radiation is studied upon the neutron and photon irradiation of composites. The characteristic X-ray radiation and conversion electrons arising from the capture of slowing neutrons of the DT reaction by liquid-crystal DNA–Gd structures are studied for their effects on biological cells. The liquid-crystal structure in the form of microparticles with a volume of $0.1 \mu\text{m}^3$ contains 10000 Gd atoms per DNA molecule. During the neutron irradiation of biological samples with a concentration of 1000 DNA–Gd particles per cell, cells are damaged by secondary radiation at a thermal neutron fluence of $\sim 10^{11} \text{ n/cm}^2$. A high-purity germanium (HPGe) spectrometer is used to study the radiation-protective properties and characteristic X-ray radiation of a Pb-containing composite upon irradiation with photons from cobalt-57 with an activity of 100 kBq.

Keywords: X-ray radiation, composite, metal, neutron, photon

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INTRODUCTION

Heavy metals are used to design radiopharmaceuticals and radioprotective materials. For example, gadolinium and ytterbium are used in medications for neutron capture therapy and the brachytherapy of malignant growths [1–4]. Lead and tungsten microparticles are applied in composite radiation-protective materials as gamma and beta absorbers [5, 6]. These elements intensively absorb photon and neutron radiation and produce X-ray radiation and conversion electrons. In case of the former, the secondary radiation from the slow-neutron capture reaction is used to damage cancer cells in a patient's body. In protective materials, gamma and beta absorption are applied to decrease the energy and penetrating power of radiation and to protect the vital organs of staff.

Gadolinium which has a large thermal-neutron capture cross section is used in composites (as a source of characteristic X-ray radiation and conversion electrons) [7]. The application efficiency of gadolinium-containing pharmaceuticals depends on the local concentration of gadolinium in these preparations. In [8–10], the properties of a liquid-crystal DNA–Gd structure with a gadolinium concentration of about 300 mg/mL were studied. The studied liquid-crystal structures with a volume of 10^{-13} cm^3 in the form of cylindrical particles contained 10^4 double-stranded DNA molecules and 10^8 Gd atoms. Three gadolinium atoms in the DNA–Gd structure were found to bind two nucleotides (phosphate groups) of the DNA mol-

ecules. The use of these composite materials as the main component of a tumor-tropic substance for neutron-radiation absorption can considerably increase the efficiency of neutron-capture therapy (NCT). A high concentration of gadolinium delivered to a tumor allows one to reduce the neutron flux required for the therapy and the harmful effect of radiation on healthy tissue.

Lead- and tungsten-containing composite materials increase several-fold the absorption of external radiation to transform it into local X-ray radiation.

METHODS

Model calculations show that the double damage of a cancer-cell nucleus results in the cell death [11]. The thermal-neutron capture reaction in DNA–Gd particles is accompanied by the formation of characteristic X-ray radiation (42.4 keV), Auger electrons (up to 7 keV), and conversion electrons (from 30 to 200 keV). Auger electrons are almost completely absorbed in DNA–Gd particles [12]. X-ray radiation and conversion electrons escape the particles and can damage the nuclei of cancer cells (Fig. 1). The probability of the formation of X-ray radiation and conversion electrons is about 50% per thermal neutron captured by a DNA–Gd particle. Biological material with a volume of 1 mL contains $\sim 10^8$ cells and, at a concentration of DNA–Gd particles of about 10^{11} cm^{-3} (or a gadolinium concentration of about 5 mg/mL^3), there can be

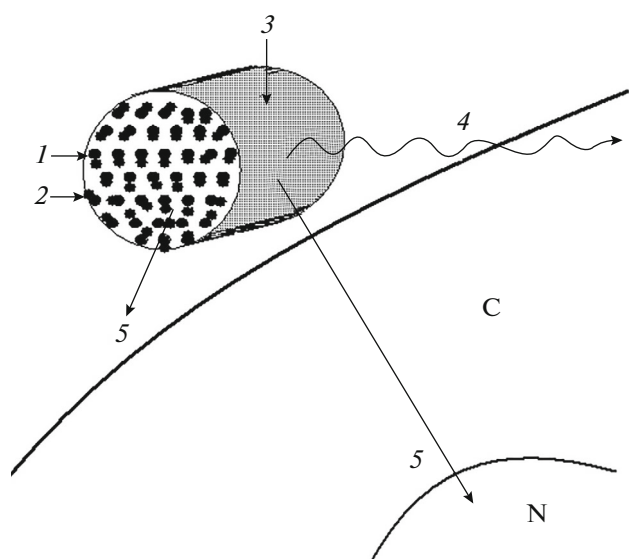


Fig. 1. Schematic diagram of neutron capture in a DNA–Gd particle with the escape of X-ray radiation, Auger electrons, and conversion electrons to the biological-cell surface. 1 is a DNA molecule, 2 is a gadolinium ion; 3 is neutron, 4 is secondary photon, 5 is secondary electron, C is a cell, and N is a cell nucleus.

the assured penetration of radiation into the cell nucleus and double damage of the cell. Double radiation capture in the DNA–Gd particles resulting in cell damage can occur at a thermal neutron fluence of $\sim 10^{11}$ n/cm².

In the present work, secondary radiation from neutron capture was studied by its action on biological cells in samples. In the neutron experiment, we used biological samples containing about 10^7 cells in a nutrient-rich aqueous solution. A single-cell layer (with a volume of 0.1 mL) was uniformly distributed over the bottom of a plastic container (Fig. 2). A layer of DNA–Gd particles (with a Gd weight of about 0.5 mg) covered the layer of biological cells and thereby created a concentration of about 1000 DNA–Gd particles per cell or a gadolinium concentration of about 5 mg/mL in the biological material.

In the study of local X-ray radiation in protective materials with lead (79 keV) and tungsten (64.5 keV), the photon spectra were measured upon the irradiation of composite materials with radiation from a cobalt-57 source. The degree of attenuation of primary photon radiation and the intensity of characteristic local X-ray radiation were determined.

EXPERIMENTAL DEVICE

A device for neutron irradiation of the biological samples was placed near the target of a neutron generator (Fig. 3). Fast (14 MeV) neutrons from the neutron generator were multiplied due to inelastic scattering and the ($n, 2n$) reaction in a tungsten converter with a

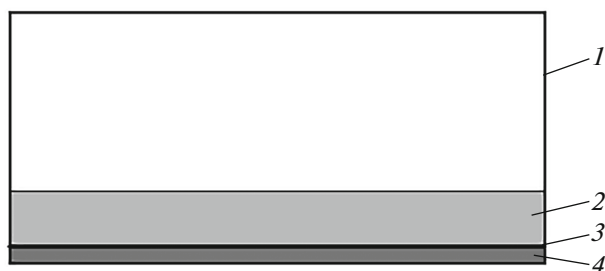


Fig. 2. Schematic diagram of the sample with biological cells and DNA–Gd particles. 1 is the container, 2 is the aqueous solution, 3 is the layer of DNA–Gd particles, and 4 is the layer of biological cells.

thickness of 2 cm and decelerated to a thermal energy in a polyethylene moderating unit with dimensions of $20 \times 20 \times 20$ cm [13]. The biological samples with cells were located inside a cube within a cavity with dimensions of $5 \times 5 \times 10$ cm. A lead reflecting converter with a thickness of 20 cm created an additional neutron flux. Bismuth and borated polyethylene layers surrounded the device to absorb external neutrons and gamma radiation.

To ensure the vital activity of cells, the temperature within the device cavity was maintained at about 36°C using a thermostat. Temperature stability was provided by adjusting the heating elements of the thermostat placed in the cavity with the biological samples and temperature gages.

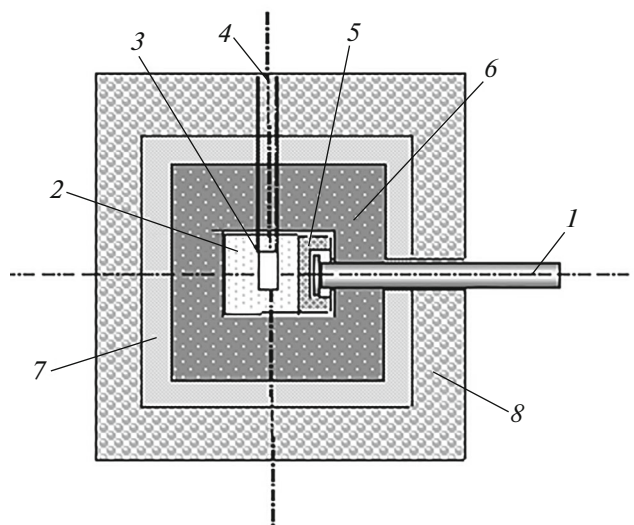


Fig. 3. Device for irradiating the biological samples. 1 is the source of fast neutrons (neutron generator), 2 is the fast-neutron moderation unit, 3 is the cavity within the cube, 4 is the inlet port for placing the samples to be irradiated, 5 is the tungsten converter, 6 is the lead reflecting converter, 7 is the layer for protection from gamma and neutron radiation, and 8 is the protective layer made of borated polyethylene.

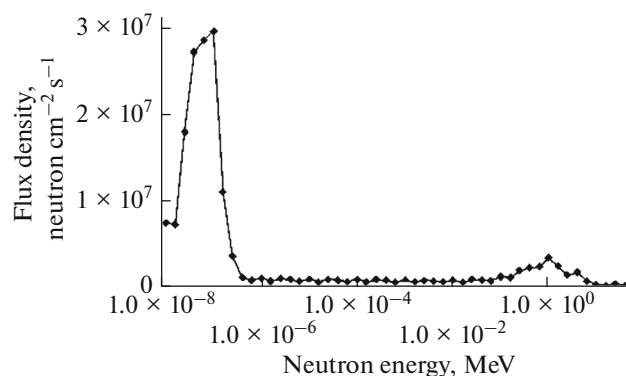


Fig. 4. Calculated energy distribution of neutrons in the cavity with irradiated biological samples inside the device.

Characteristic X-ray radiation in the protective composite was studied using a cobalt-57 source with an activity of 10^5 Bq and a photon energy of 122.06 keV (85.5%) and 136.47 keV (10.7%). The spectra of X-ray radiation from the protective material and photons from the source were measured using a low-background gamma spectrometer based on a high-purity germanium detector [14].

RESULTS AND DISCUSSION

To optimize the experimental conditions, slow and fast neutron fluxes in the moderator were measured and calculated. Calculation was performed using the NCNP4B program and the MCNPXS library. Measurements were performed by activation analysis using Mn, Nb, and In samples. The values of slow and fast neutron fluxes at the site of the biological samples are given in Fig. 4.

The samples with biological cells were irradiated in several sessions. Prior to each session, three cell samples, viz., a control sample, a sample with DNA-Gd particles, and a sample free of particles, were prepared. The irradiation time for each of two samples was from 15 min to 1 h. The fluence of thermal and fast neutrons was measured by the activation method and varied in the range of $(0.5-5) \times 10^{11}$ n/cm² depending on the deuteron current across the NG-400 target. The fast- and thermal-neutron flux density in the samples varied in the ranges of $(1-3) \times 10^7$ and $(0.5-1.5) \times 10^8$ n/cm²/s, respectively.

After the sessions, the samples were stored for several days and checked for the presence of living proliferating cells. The absence of such cells in a sample suggests cell death under irradiation.

The studies showed that, after the session with a thermal-neutron fluence of above 10^{11} n/cm², the cells in the biological samples containing DNA-Gd particles died and did not proliferate (Fig. 5). The biological cells in the control samples and the irradiated samples free of DNA-Gd particles remained alive and proliferated. This suggests that additional characteris-

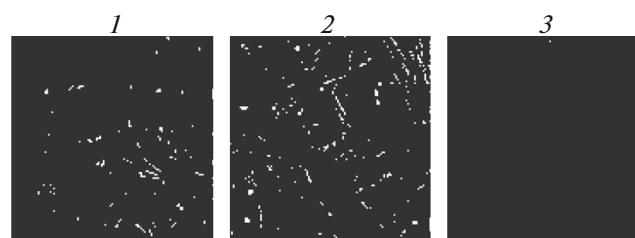


Fig. 5. Optical image of biological cells from the samples after irradiation (obtained using a Leica DMI4000 microscope); 1 is the unirradiated control sample, 2 is the neutron irradiated sample free of DNA-Gd particles (open cell points), and 3 is the irradiated sample with DNA-Gd particles.

tic X-ray radiation and conversion electrons from neutron capture in DNA-Gd particles cause damage to the biological cells. The studied composite material for neutron-capture therapy has a high concentration of gadolinium and allows one to decrease several fold the required neutron flux and the harmful effect of irradiation.

Upon gamma irradiation of the radiation-protective material, the spectra of characteristic X-ray radiation and the spectra of attenuated gamma radiation of

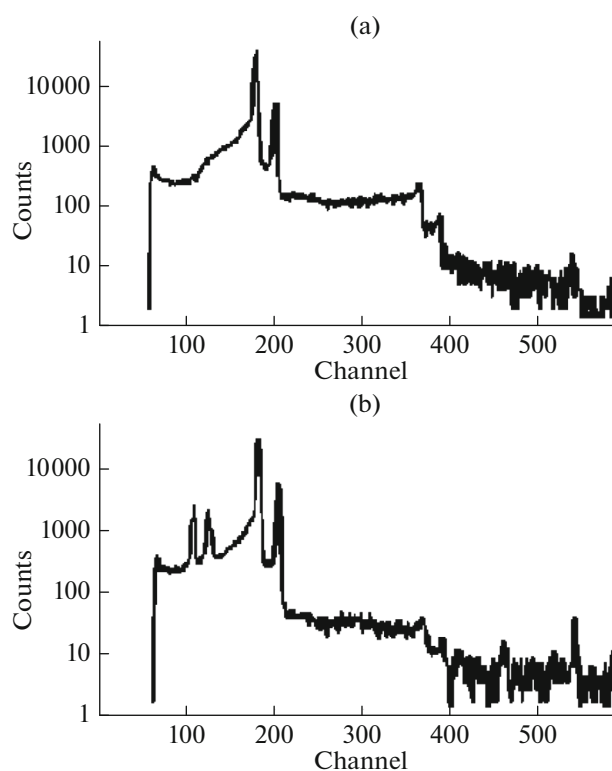


Fig. 6. Spectra of photons from the ⁵⁷Co source measured on a low-background gamma spectrometer: (a) spectrum of photons from an open ⁵⁷Co source; and (b) spectrum of photons from a ⁵⁷Co source shielded by protective material.

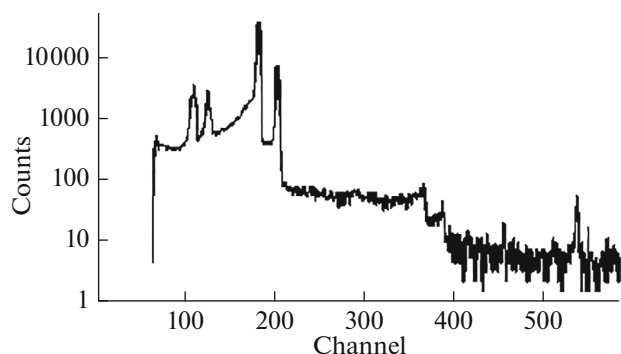


Fig. 7. Spectrum of secondary X-ray radiation and photons from the ^{57}Co source shielded by a lead plate.

the ^{57}Co source transmitted through the metal composite were measured (Fig. 6). The spectrum from an open ^{57}Co source was measured for 50 s. The spectrum of the ^{57}Co source shielded by the protective material was measured for 57 s. A peak in the spectrum of the characteristic X-ray radiation at an energy of about 80 keV indicates the presence of a lead component in the protective material. The characteristic X-ray radiation intensity was 3% of the intensity of the source line at 122 keV. The presence of lead elements in the protective composite was confirmed by measuring the spectrum of characteristic radiation from the lead plate shielding the source (Fig. 7). The analysis of gamma peaks on the spectra at energies of 122.06 and 136.47 keV allowed us to determine the values of gamma-radiation attenuation by the protective material at these energies, which were 3.7 and 2.8, respectively. The concentration of the lead component in the protective material was estimated from these values to be about 0.3 g/cm^2 .

CONCLUSIONS

Studies of heavy metal-containing composites are of significance for the designing nuclear medicines and radiation-protective materials. Gamma and beta radiation and neutrons (as the most common and pen-

etrating types of radiation) can be absorbed effectively by the components of composite materials and can create local X-ray and electron radiation. Data from the studies performed are important for designing composite materials with a high radiation absorptivity and for decreasing the harmful effect of radiation on humans.

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