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INTERACTION OF FLUXES OF FAST AND THERMAL NEUTRONS WITH AN AQUEOUS SOLUTION OF ORGANIC DYE METHYLENE BLUE CONTAINING AND NOT CONTAINING BORIC ACID

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The processes of interaction of neutron fluxes with an aqueous solution of the organic dye methylene blue (MB) – $C_{16}H_{18}N_3SCl$ containing and not containing 4% boric acid (H_3BO_3) were studied. The work was carried out at the linear electron accelerator LUE-300 NSC KIPT. A set of tungsten plates was used as a neutron producing target. The electron energy was 15 MeV, the average current was 20 μA. The samples were behind lead shielding and without it, with and without a moderator. The energy spectra of neutron fluxes are calculated at the location of samples with a moderator with a thickness of 0 to 5 cm. The total neutron fluence was $2 \cdot 10^{11}$ n/cm^2.

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INTRODUCTION

The huge layers of knowledge accumulated by humanity in the course of comprehensive research in the field of the nucleus provide little limited opportunities for the modern industrial-technological order. Industries such as materials science, nuclear energy, nuclear medicine, nuclear chemistry, nuclear biology, etc. actively use the possibilities of the impact of ionizing flows on matter, which in due time actualizes the problem of registering these flows. Along with such methods of measuring the absorbed dose as ionization, optical, luminescent, calorimetric, etc. [1-3], chemical methods of dosimetry [4] are preferable due to their sufficient availability and simple methods of use. Research in this area is being intensively conducted in many research centers around the world.

The work was carried out in order to study the processes of interaction of neutrons with an aqueous solution of the organic dye methylene blue (MB) – $C_{16}H_{18}N_3SCl$, with the prospect of using this dye in a system for registering thermal and epithermal neutron fluxes.

The main property of dyes is the ability to intensively absorb and convert light energy in a certain region of the spectrum [5]. The area of practical application of the dye depends on the nature of the transformation of the absorbed energy [6]. Previous studies [7-9] have shown that aqueous solutions of organic dyes are more sensitive to ionizing radiation, as a result of their oxidation by radicals and radical ions formed during the radiolysis of solvents ($OH^{\cdot}$, $HO_2^{\cdot}$ and etc.). When exposed to ionizing radiation on a solution of an organic dye, an irreversible loss of color occurs (a decrease in the intensity of the long-wavelength band of the absorption spectrum) of the solution. The fact that an organic dye solution has intense absorption and fluorescence bands in the visible region of the spectrum gives grounds for the development and creation of a chemical system for recording the total absorbed dose on its basis. For example, aqueous solutions of dyes can be used to determine the radiation dose in the range of 0.003…0.5 Mrad, and polymer films dyed with dyes – 0.3…40 Mrad.

OBJECTS OF RESEARCH

This research work was carried out at the Kharkov Institute of Physics and Technology (KIPT) utilizing the Linear Accelerator LUE – 300. The processes of interaction of neutron fluxes with an aqueous solution of the organic dye methylene blue (MB) containing and not containing 4% boric acid, using a moderator with a thickness of 0 to 5cm, as well as with and without lead shielding, were the subject of research in this work.

In researches related to the registration of neutron fluxes, the use of the following nuclear reactions is most common: ^{10}B (n, α) ^{7}Li, ^{6}Li (n, α) T, ^{3}He (n, p) T, elastic scattering, inelastic scattering [10, 11]. Elastic neutron scattering and the nuclear reaction ^{10}B (n, α) ^{7}Li, were used in these studies.

Elastic scattering of neutrons

The process of elastic neutron scattering can proceed in two ways:

***– potential scattering*;**

– elastic resonant scattering.

Elastic-resonant scattering (*elastic scattering through the formation of a compound nucleus*) is a neutron scattering reaction, as a result of which the compound nucleus, upon transition to the ground state, emits a neutron with a kinetic energy less than the kinetic energy of the neutron before colliding with the nucleus [12, 13].

Potential scattering – neutron scattering on the nucleus as on an elastic body, i.e. elastic scattering in the field of nuclear forces. Interaction of this type is accompanied only by the redistribution of kinetic energy and momentum between the neutron and the target nucleus [12, 13].

Elastic neutron scattering is used to detect fast neutrons by observing traces of recoil nuclei in various track detectors, to measure their kinetic energy, and also to detect recoil nuclei by ionization methods. Elastic scattering of neutrons by hydrogen nuclei is used to detect fast neutrons by recoil protons. The mass of a proton is close to the mass of a neutron. The neutron loses its kinetic energy in a central collision with a hydrogen atom (proton).

In view of the fact that an aqueous solution of the dye was studied in this work, the loss of color was due to two factors:

– *radiolysis of water as a result of oxidation by radicals ($OH^{\cdot}$, $HO_2^{\cdot}$, etc.);*

– *kinematic disintegration of dye molecules upon collision with fast neutrons.*

Neutron absorption reaction ^{10}B (n, α) ^{7}Li

The scheme of the nuclear reaction of absorption of a ^{10}B neutron is shown in Fig. 1. Science knows 14 isotopes of boron, of which only two are stable: ^{10}B and ^{11}B [14] with a content in nature of 18.2 and 81.8%, respectively. Due to the very large absorption cross section of thermal neutrons by the ^{10}B isotope (3837 ± 9 barn), their intense absorption occurs [15], which leads to an instantaneous nuclear reaction ^{10}B (n, α), which results in the formation of an α particle and a ^{7}Li nucleus. This reaction proceeds through two channels:

$^{10}B + n \rightarrow (^{7}Li)^{*} + \alpha + E_1$;

$^{10}B + n \rightarrow {}^{7}Li^{*} + \alpha + E_2$,

where E_1 and E_2 are the reaction energy, is released in the form of the kinetic energy of the reaction products. In the first channel, the ^{7}Li nucleus is formed in an excited state with an excitation energy of 0.48 MeV. The excited state of ^{7}Li is marked with an asterisk. The transition of the nucleus from the excited state to the ground state is accompanied by the emission of a γ-quantum with an energy of $E\gamma$ = 0.48 MeV. Therefore, the first reaction channel can be rewritten as follows:

$$^{10}B + n \rightarrow {}^{7}Li + \alpha + E_\gamma + E_1$$

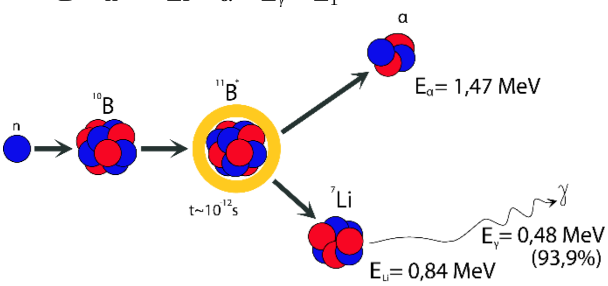

Fig. 1. Scheme of the nuclear reaction of neutron absorption by ^{10}B

The reaction energy ^{10}B (n, α) ^{7}Li is 2.79 MeV. One part of the energy (E_γ=0.48 MeV) is carried away by the γ-quantum, the other part (E_1=2.31 MeV) is released in the form of the kinetic energy of the α-particle and the nucleus of lithium. At the same time, from the energy E_1=2.31 MeV, the share of the α-particle is E_α=1.47 MeV, and the share of the lithium nucleus is E_{Li}=0.84 MeV. The probability of the reaction proceeding through the first channel is 0.939 for slow neutrons with an energy of about 10 keV. Then this probability gradually decreases, reaching a value of 0.3 at a neutron energy of 1.8 MeV and again increasing to 0.5 at a neutron energy of 2.5 MeV. Through the second channel, the reaction ^{10}B (n, α) ^{7}Li proceeds with a probability of 6.1% for slow neutrons and, accordingly, with a higher probability for fast neutrons. The reaction energy E_2 = 2.79 MeV in this case is completely transferred to the α-particle and the lithium nucleus.

Thus, when boric acid is added to a dye solution and it is irradiated with a thermal neutron flux, the above reactions occur. At the same time, the interaction of high-energy α-particles and the ^{7}Li nucleus with dye molecules leads to their disintegration, and the presence of a gamma quantum in the first reaction leads to water radiolysis and ionization of atoms in molecules.

EXPERIMENTAL PART

A schematic diagram of the experimental setup, utilized for investigating the interaction of neutron fluxes with an aqueous solution of methylene blue (MB) –$C_{16}H_{18}N_3SCl$, with and without 4% boric acid (H_3BO_3), is depicted in Fig. 2. An electron beam with an energy of 15 MeV and an average current of 20 μA was output from a linear accelerator LUE – 300 to a neutron-producing target, which was located at a distance of 40 cm from the output foil. The neutron producing target was a set of tungsten plates. From lead 5 cm thick, protection against scattered electrons and the accompanying gamma background was assembled in the immediate vicinity of the target. The dye solutions in glass test tubes were placed inside a lead shielding at a distance of 15 cm from the electron beam axis. As a moderator, 5 cm thick polyethylene was placed between the lead shielding and the test tubes. The neutron-producing target, as well as the entire structure, was intensively blown with air to prevent overheating. The total neutron fluence from the converter was $2 \cdot 10^{11}$ n/cm^2. The change in the dye concentration as a result of irradiation was determined from the optical absorption spectra.

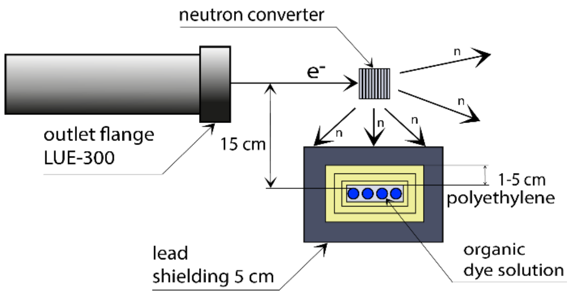

Fig. 2. Scheme of an experiment on target irradiation with a neutron flux

Fig. 3 shows a photograph of the experimental design on which the studies were carried out. This design was located at the output of LINAC LUE – 300, the photo also shows the location of the neutron generator and the lead shielding inside which the test samples were located.

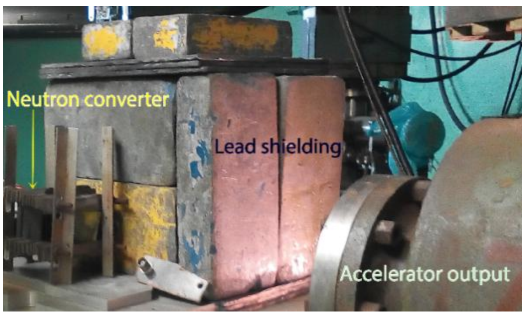

Fig. 3. Experimental construct on which the research was carried out

The energy spectra of neutron fluxes inside the lead shielding at the location of the experimental samples were calculated in order to obtain the most complete picture of the effect of neutrons on a solution of the organic dye methylene blue (MB). The GEANT-4 software code [16] was used for modeling. Fig. 4 shows the calculation results. Six options were considered in the calculations: 0 – neutron spectrum inside the shielding at the location of the sample without a polyethylene moderator, 1–5 – neutron spectra inside the shielding with a gradual increase in the thickness of the moderator from 1 to 5 cm. When using a moderator 5 cm thick, the number of fast neutrons is reduced by a factor of 10 compared to the number of neutrons when there is no moderator. At the same time, the number of thermal neutrons increases by about 2 times in comparison with the number of thermal neutrons without a moderator. The total neutron fluence for the case without a moderator at the location of the samples for the presented experiment was 10^{11} n/cm^2.

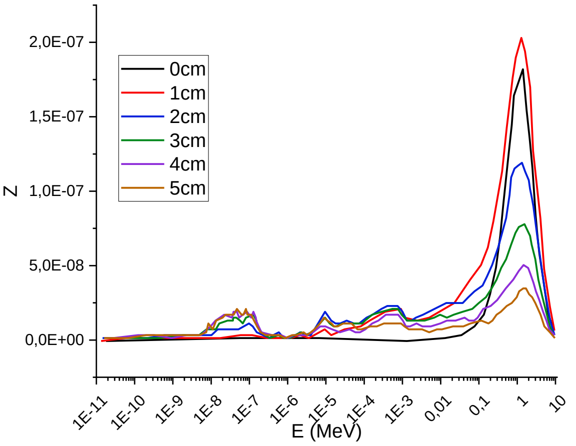

Fig. 4. Energy spectra of neutron fluxes of a neutron generator (normalized to 1 electron with an energy of 15 MeV) without a moderator and with a moderator of various thicknesses

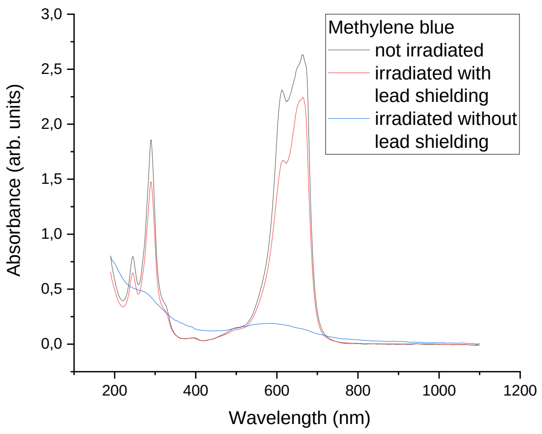

Fig. 5. The main absorption spectra of the irradiated and non-irradiated dye not containing boric acid: black curve - solution before irradiation; red curve - solution irradiated behind a lead shielding; the blue curve is the solution irradiated outside the lead shielding

In order to make the study as complete as possible, a number of experiments were carried out. An aqueous solution of the organic dye methylene blue (MB) was prepared in two versions: with and without 4% boric acid (H_3BO_3). In the initial series of experiments, test tubes with these aqueous solutions were placed inside and outside the lead shielding at a distance of 15 cm from the neutron generating target. The main absorption spectra of the irradiated and non-irradiated dye without the addition of boric acid are shown in Fig. 5.

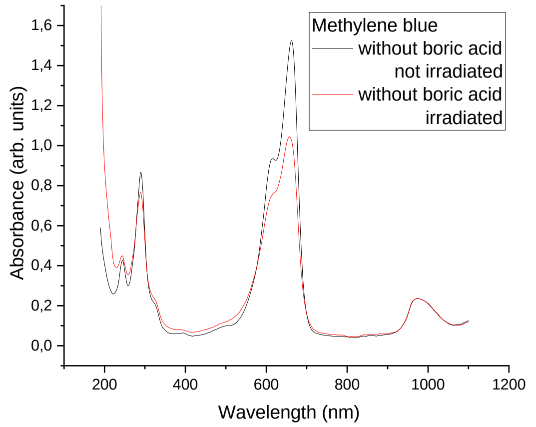

Fig. 6. Absorption spectra of the irradiated and non-irradiated dye not containing boric acid by neutron fluxes in the presence of a 5 cm thick polyethylene moderator

As can be seen in Fig. 5, the dye molecules are completely destroyed (the dye loses color) when the solution is irradiated without a moderator and without lead shielding. The loss of color of the dye solution occurs mainly as a result of the effect of scattered electrons and gamma rays on it. The same figure shows that lead protection is very effective, because. the loss of color of the MW dye when it is irradiated behind protection is 10%. The destruction of dye molecules in this case occurs as a result of their interaction with fast neutron fluxes.

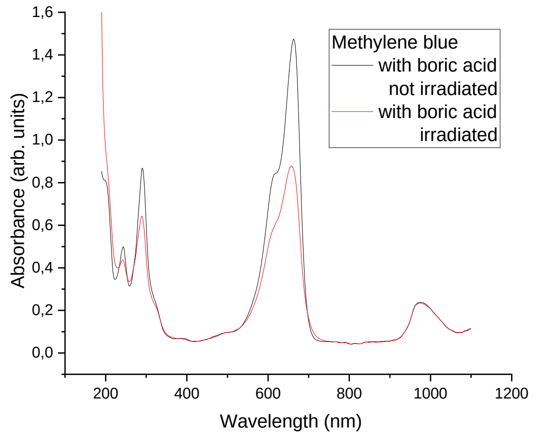

Fig. 7. Absorption spectra of a dye containing 4% boric acid irradiated and non-irradiated by a neutron flux using a 5 cm thick polyethylene moderator

In the next series of experiments, one tube with an aqueous solution of an organic dye containing boric acid and a second tube without boric acid were placed inside a lead shielding. A moderator 1 to 5 cm thick was placed between the test tubes with the objects of study and the lead shielding. At the same time, the geometric parameters of the experiment were preserved, the distance from the test tubes to the neutron generator was also 15 cm. The main absorption spectra of the irradiated and non-irradiated dye in the presence of a moderator 5 cm thick: without boric acid are shown in Fig. 6, and with a content of four percent boric acid in Fig. 7.

When a moderator made of polyethylene 5 cm thick is placed between the lead shielding and the irradiated sample that does not contain 4% boric acid, there is (see Fig. 6) a 15% destruction of the MB dye molecules is observed as a result of the action to thermal neutrons on it. With the addition of 4% boric acid to the MB dye solution and the placement of a polyethylene moderator 5 cm thick between the lead shielding and the irradiated sample, the picture changes significantly (see Fig. 7), namely: 26% destruction of the dye molecules is observed as a result of the exothermic reaction absorption of the neutron ^{10}B (n, α) ^{7}Li.

CONCLUSIONS

The processes of interaction of neutron fluxes with an aqueous solution of the organic dye methylene blue (MB) – $C_{16}H_{18}ClN_3S$ with and without 4% boric acid – H_3BO_3 content were studied in this work. The samples under study were subjected to neutron irradiation, while they were placed behind the lead shielding and outside it, with and without a moderator.

The energy spectra of neutron fluxes at the location of the experimental objects were calculated using the GEANT-4 program code for this experiment.

An analysis of the experimental data showed that when objects are irradiated without lead shielding and a moderator, dye molecules are completely destroyed. The destruction of the dye molecules, in this case, occurs mainly as a result of its interaction with scattered electrons and gamma rays. A 10% destruction of MB was observed as a result of the interaction of dye molecules with fast neutron fluxes using a 5 cm thick lead shielding. A 15% destruction of dye molecules without boric acid on thermal neutrons was observed when a 5 cm polyethylene moderator was installed between the lead shielding and the object of study. The interaction of thermal and epithermal neutron fluxes with an aqueous solution of the organic dye methylene blue (MB) – $C_{16}H_{18}ClN_3S$ with a 4% content of boric acid leads to 26% destruction of the dye molecules as a result of the nuclear reaction of neutron absorption ^{10}B (n, α) ^{7}Li.

The works performed has shown that an aqueous solution of the organic dye MB is a convenient object for studying the processes of interaction of ionizing radiation with matter. MB can also be a good material for creating detectors of ionizing radiation, in particular systems for chemical registration of thermal and epithermal neutron fluxes. Such detectors can be used for radioecological monitoring of the environment, in nuclear power engineering and nuclear medicine.

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ВЗАЄМОДІЯ ПОТОКІВ ШВИДКИХ І ТЕПЛОВИХ НЕЙТРОНІВ З ВОДНИМ РОЗЧИНОМ ОРГАНІЧНОГО БАРВНИКА МЕТИЛЕНОВИЙ СИНІЙ, ЯКИЙ МІСТИТЬ І НЕ МІСТИТЬ БОРНУ КИСЛОТУ

В.В. Цяцько, С.П. Гоков, Ю.Г. Казаринов

Досліджено процеси взаємодії потоків нейтронів з водним розчином органічного барвника метиленовий синій (МС) – $C_{16}H_{18}N_3SCl$, що містить і не містить 4% борної кислоти (H_3BO_3). Робота виконувалася на лінійному прискорювачі електронів ЛУЕ-300 ННЦ ХФТІ. Як нейтронопродукуюча мішень використовувався набір вольфрамових пластин. Енергія електронів становила 15 МеВ, середній струм – 20 мкА. Зразки знаходилися за свинцевим захистом і без нього з отеплювачем і без. Розраховані енергетичні спектри потоків нейтронів у місці розташування зразків із сповільнювачем товщиною від 0 до 5 см. Сумарний флюєнс нейтронів становив $2 \cdot 10^{11}$ н/см^2.