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INVESTIGATION OF INTERACTION MECHANISMS OF HIGH ENERGY ELECTRONS AND GAMMA QUANTUM WITH AQUEOUS SOLUTION OF METHYL ORANGE DYE[†]

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The level of development of modern nuclear technologies forms a request for the development of new branches of science. At the same time, chemical dosimetry methods are also being improved [1, 2]. The essence of such methods consists in the quantitative determination of the radiation-chemical damages to the molecules of a substance when it is exposed to ionizing radiation [3, 4]. Liquid and solid solutions of organic dyes have intense bands optical absorption and fluorescence in the visible region of the spectrum, which makes it possible to use them in dosimetry systems [5, 6]. The use of organic dyes makes it possible to determine the absorbed dose in the range from 10^{-6} to 10^{4} M Rad [7, 8]. In this work, we studied the processes of interaction of gamma-ray and high-energy electron fluxes with an aqueous solution of the organic dye methyl orange ($C_{14}H_{14}N_3O_3SNa$) [9, 10]. The calculations and experiment were carried out on a resonant electron accelerator with energies up to 30 MeV. The electron beam energy was 15 MeV. A tungsten converter was used to generate gamma quanta. The thickness of the converter varied from 0 to 6 mm. We have developed a computer program in C++ to simulate the irradiation process. This program uses the Geant4 class library based on the Monte Carlo method and runs in multi-threaded mode. For calculations, the model "PhysicsList emstandard_opt3" was chosen as the most suitable one. The value of radiation damage per one incident electron and produced gamma-quantum is determined in the work. The simulation results are compared with experimental data. Based on the results obtained, conclusions were drawn about the main mechanisms leading to the decomposition of organic dye molecules, and methods for optimizing the experiment for further research were proposed.
Keywords: organic dye, electron, gamma quantum, dosimetry
PACS: 61.72.Cc, 61.80.Hg, 78.20.Ci, 87.80.+s, 87.90.+y, 07.05.Tp , 78.70.−g

Soon after the discovery of X-ray radiation, the need arose for scientific research on the processes of interaction of ionizing radiation with complex organic objects. The result of such research can be the solution of many problems in various areas of science, as well as many applied problems.

The current level of development of nuclear energy and nuclear technologies is such that there is a need for new approaches to the organization of radiation protection. There is also a need for modern, more compact, easy-to-handle and competitive dosimetry systems and methods. The development of industrial capacities and space exploration generates a need for new materials with modified molecular structures that have both resistant properties to radiation and increased strength characteristics. The results obtained in the course of studies of the mechanisms of interaction of ionizing radiation with molecules of organic matter can be completely extrapolated to a living organism. A living organism is a complex organic system with a number of different properties. These properties can be attributed to both biological and chemical, and physical [11]. Thus, the interest of medical science in such research becomes obvious. For medicine in general and for radiation therapy in particular, organic dyes can be a good research material [12]. This is not a complete list of problems that can be solved by the research that we are conducting in the framework of this work.

In this work, research was carried out with the aim of developing a methodology for using the degradation of organic dye molecules that have been exposed to ionizing radiation as a tool for determining the absorbed dose.

COMPUTER SIMULATION OF EXPERIMENTAL CONDITIONS

Real experiments are planned to study the radiation damage processes of a methyl orange organic dye solution under the influence of an electron beam. The energy of the incident electrons is 15 MeV in an experiment. Before carrying out a series of experiments, it is necessary to carry out computer simulation in order to preliminary estimate the experimental conditions and expected results. Changes to the planned conditions of the experiment are possible based on the preliminary calculations results, as well as geometric parameters clarifications. These parameters include thickness and quantity of tungsten converter layers, distance to the target, etc. Therefore, the estimation of some important parameters required computer simulation. A computer program has been developed in the C++ object-oriented programming language to solve this task. Nuclear-physical processes that occur during the passage of radiation through matter are described in this work using the Geant4 class library [13]. The Geant4 library is a modern toolkit that

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is essential for solving practical problems to describe linear electron accelerators models. In particular, one can solve the tasks of developing and improving the parameters of bremsstrahlung converters using Geant4 toolkit.

The program we have developed contains several main modules. There are modules for describing the geometry of an experiment, a module for describing the primary particles source, a module for determining models of physical processes, etc. In addition, the program contains some functions required to select the appropriate level of the results detail.

The Geant4 toolkit is based on the Monte Carlo method. Therefore, it is necessary to simulate a large number of primary particles passing through the experimental setup to obtain results with a minimum statistical error in calculations. The quantity of primary electrons $N_e=10^7$ for performing virtual nuclear physics experiments. The statistical error of calculation by the Monte Carlo method is less than 1% in this case. The use of a large number of primary particles usually leads to a calculation time increasing. The calculations were carried out in a multithreaded mode in order to reduce the total computation time. These calculations were performed using the OpenMPI library [14], which is free and open source software.

The module for describing physical processes in the developed program is based on the “emstandard_opt3” model, which is most suitable [15, 16] in the considered energy range (up to 15 MeV) for all primary and secondary particles. The threshold energies E_{cut} were chosen to be equivalent to a range of 100 µm in the substance. The user does not directly define the energy threshold because there is a special method of threshold calculations [15] used in Geant4 for different materials. The user defines a unique cut in range [13], whose value is transformed into a kinetic energy threshold per each material at initialization time of user’s program.

The schematic diagram of the experimental setup is shown in Figure 1. Elements of this scheme were used to describe the model geometric parameters used in the developed program.

The distance between the accelerator outlet window (Figure 1) and the bremsstrahlung converter is 50 mm. The accelerator outlet window contains titanium foil. The thickness of the foil is 0.05 mm. The real converter is made of tungsten. Transverse dimensions of the converter are 50 mm×50 mm. The target transverse dimensions are 10 mm×10 mm. The thickness of the converter was varied from 1 to 8 mm in this series of computational experiments. These calculations are necessary to determine the thickness of the converter that produces the maximum amount of gamma quanta that reach the target boundaries.

The simulated energy spectra of bremsstrahlung gamma quanta emitted from the tungsten converter and reached the target are shown in Figure 2 for different values of the converter thickness. The results are normalized to 1 incident electron.

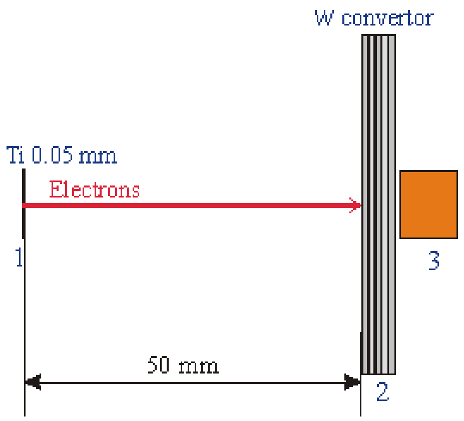

Figure 1. Simplified diagram of the experimental setup: 1 – accelerator outlet window; 2 – bremsstrahlung converter; 3 – the target

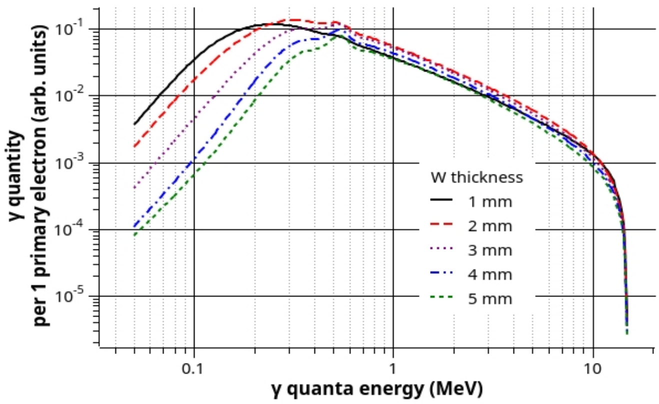

Figure 2. Bremsstrahlung gamma quanta energy spectra for different thickness values of the tungsten converter

As a result of the calculation data analysis, it turned out that the maximum yield of bremsstrahlung gamma quanta is at a tungsten plate thickness of 2 mm. The amount of bremsstrahlung gamma quanta in front of the target slightly decreases at a plate thickness of 3 mm.

The presented materials belong to a series of our works on the study of the behavior of organic substances under the action of ionizing radiation. We carried out measurements with a solution of the organic dye methylene blue ($C_{16}H_{18}N_3SCl$) in a previous publication [17]. The results are shown in Figure 3 and Figure 4 for various converter thickness values. The result of determining the most probable values of the energy absorbed in the target with methyl orange dye solution is shown in Figure 5. The target dimensions are 10 mm ×10 mm × 10 mm. The most probable value of the energy absorbed in the solution is 1.65 MeV for incident electrons with an initial energy of 15 MeV.

Therefore, it is expedient for us to study the processes of the organic dye destruction for several thickness values of the bremsstrahlung converter. We chose 2 mm, 4 mm, and 6 mm. The flow of particles hitting the target contains mainly gamma quanta at 4 mm converter thickness. The flow of particles crossing the target boundaries contains a negligibly small number of electrons at a converter thickness of 6 mm due to the complete stoppage of primary electrons in the converter material. The flow of particles hitting the target contains gamma quanta, as well as electrons, at 2 mm converter thickness. These values chosen by us are sufficient for studying the dynamics of the processes that occur in the substance of the methyl orange dye under the influence of ionizing radiation.

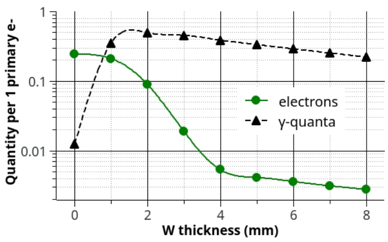

Figure 3. The bremsstrahlung gamma quanta flux in comparison with the electrons flux [17] in front of the target

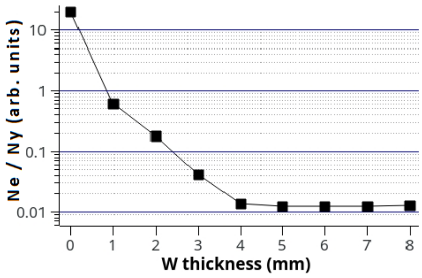

Figure 4. Coefficient of the ratio of electrons and gamma quanta fluxes [17] in front of the target

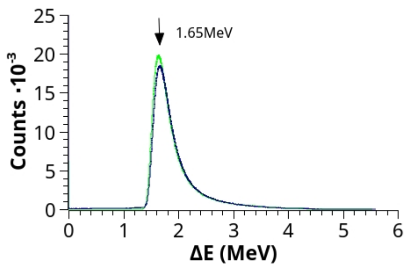

Figure 5. The most probable value of the energy absorbed in the solution for 15 MeV incident electrons

DESCRIPTION OF THE EXPERIMENT

The experimental part of the work was carried out on the LINAC LUE-300 NSC KIPT [12, 17]. The target was an aqueous solution of methyl orange dye with a volume of 1 cm^3. It was located in the path of the electron beam in a rigidly fixed glass tube. Target positioning accuracy was ensured by using a fixed stand. This made it possible to set the test tube in a constant position when changing solutions relative to the axis of the accelerator. Conventionally, the experiment can be divided into two parts: irradiation of the target with an electron beam and irradiation of the target with a beam of gamma rays. To generate gamma quanta, a converter was used, which consisted of a set of tungsten plates 50 × 50 × 2 mm in size. The converter was located in front of the target in the direction of the beam. The total thickness of the converter was changed during the experiment in increments of 2 mm (0, 2, 4, and 6 mm).

A tungsten converter, when irradiated with an electron beam, is also a source of neutrons. However, the contribution of neutrons to the change in the color of the dye solution does not exceed ten percent [12]. Since the destruction of methyl orange molecules under the action of a neutron flux on it is insignificant, this contribution was not taken into account in the work.

The experiment was carried out with an electron beam energy of 15 MeV. The current density in this case was 1 $\mu A/cm^2$.

The equipment on which the experiments were carried out is shown in Figure 6.

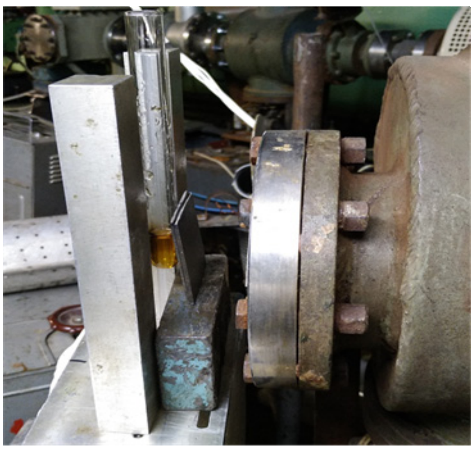

Figure 6. Experimental setup

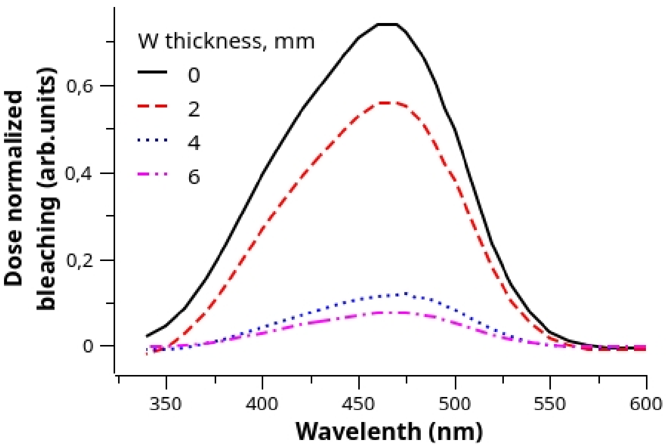

Figure 7. Degradation of methyl orange under the influence of ionizing radiation (normalized to 1 second)

During this experiment, considerable attention was paid to the concentration of the dye solution. To obtain the minimum error in determining the change in the optical density of the solution under the action of irradiation, the initial concentration of the dye in the solution was chosen so that the optical absorption at the maximum of its absorption band (460 nm) was close to unity.

Figure 7 shows the differential absorption spectra (before and after irradiation) normalized per unit time (dose) of irradiation. The spectra were measured on a single-beam spectrophotometer SF-46, in the range from 300 to 600 nm, since the main optical absorption peak of an aqueous solution of methyl orange is in this region. As can be seen from the graph, when exposed to ionizing radiation for one second on an aqueous solution of methyl orange, a significant destruction of the dye molecules occurs. A decrease in the degree of degradation of dye molecules is observed with an increase in the thickness of the tungsten converter. The black solid line in Figure 7 shows the degree of dye destruction when exposed to an electron beam, that is, at zero tungsten thickness.

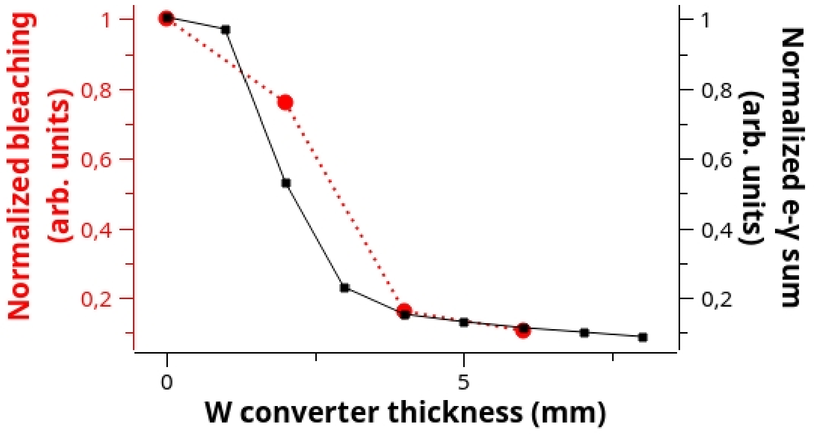

Figure 8. Normalized radiation destruction of the methyl orange solution – left curve, the normalized sum of the number of electrons and gamma quanta that affected the methyl orange solution – right curve

Using the dependences of the number of electrons and gamma quanta that interacted with the dye solution (the dependences are shown in Figure 3), an analysis was made of the efficiency of dye destruction depending on the thickness of the tungsten converter. This analysis was performed by comparing the change in optical absorption at the wavelength maximum (460 nm) (left dependence in Figure 8) and the normalized sum of the number of electrons and gamma quanta (right dependence in Figure 8). During the analysis, it was found that at a ratio of electrons and gamma quanta equal to 1/12, the maximum coincidence of the dependences was obtained.

When the radiation length of an electron in tungsten is 3.5 mm, the maximum thickness of the converter used does not exceed two radiation lengths. From here, it follows that the average energy of electrons in front of the target is about 2 MeV or more, which exceeds the average value of the electron energy loss in the solution equal to 1.65 MeV. In this case, the influence of the thickness of the converter on the loss of color of the solution when exposed to electrons can be reduced only to a change in the number of electrons. The dependence of the spectrum of gamma rays on the thickness of tungsten is more pronounced in the energy range up to 0.5 MeV (see Figure 2). In this range, the photoelectric effect, that is, ionization prevails in the interaction cross section. On the other hand, the fraction of gamma quanta with an energy of more than 0.5 MeV does not change significantly, but the Compton effect becomes the dominant interaction channel, and this ensures the destruction of the dye when using converters with a thickness of 4 and 6 mm, when the number of electrons, compared with the initial flow, decreases by 40 times or more, and the level of loss of color by the solution is only 6-8 times.

CONCLUSIONS

In this work, a research was carried out of the processes that occur when an aqueous solution of an organic dye methyl orange ($C_{14}H_{14}N_3O_3SNa$) is exposed to flows gamma quanta and electron. The gamma-ray flux was generated by a relativistic electron beam with an energy of 15 MeV, using a tungsten bremsstrahlung converter of various thicknesses.

In this work, we compared the results of computer simulation of the processes that occur during the passage of an electron beam through a tungsten converter of different thicknesses (0, 2, 4, 6 mm) with the data on the degradation of the dye solution, which were obtained during the experimental. This made it possible to separate the effects of the interaction of electrons from the effects of the interaction of gamma quanta on the dye solution.

We also carried out an approximation of the experimental dependence of the efficiency of the loss of color of the methyl orange solution on the thickness of the tungsten converter. As a result, it was found that the effect of relativistic electrons on the dye leads to the destruction of its molecules 12 times more efficiently than the effect of bremsstrahlung gamma-ray fluxes.

Since the difference in the effectiveness of the effect of these types of ionizing radiation on the methyl orange dye solution is so significant(1/12), we can state the following: when determining the absorbed dose of electron irradiation with an error of less than 10%, the accompanying gamma background can be neglected, provided that the quantitative ratio of electrons and gamma quanta is one.

Based on the studies we have done in this work, it can be argued that an aqueous solution of the organic dye methyl orange is the most optimal object for measuring the absorbed dose of a substance.

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ДОСЛІДЖЕННЯ МЕХАНІЗМІВ ВЗАЄМОДІЇ ВИСОКОЕНЕРГЕТИЧНИХ ЕЛЕКТРОНІВ І ГАММА-КВАНТІВ З ВОДНИМ РОЗЧИНОМ БАРВНИКА МЕТИЛОВОГО ОРАНЖОВОГО

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Рівень розвитку сучасних ядерних технологій формує запит у розвиток нових галузей науки. Натомість удосконалюються і хімічні методи дозиметрії [1, 2]. Суть таких методів полягає у кількісному визначенні радіаційно-хімічних ушкоджень молекул речовини при впливі на нього іонізуючого випромінювання [3, 4]. Рідкі та тверді розчини органічних барвників мають інтенсивні смуги поглинання та флуоресценції у видимій частині спектру, внаслідок чого можуть використовуватись у системах дозиметрії [5, 6]. Використання органічних барвників дозволяє визначати поглинену дозу в діапазоні від 10^{-6} до 10^{4} Мрд [7, 8,]. У цій роботі досліджувалися процеси взаємодії потоків гамма-квантів та високоенергетичних електронів з водним розчином органічного барвника метиловий оранжевий ($C_{14}H_{14}N_3O_3SNa$) [9, 10]. Розрахунки та експеримент було проведено на резонансному прискорювачі електронів з енергією до 30 МеВ. Енергія електронного пучка становила 15 МеВ. Для генерації гамма-квантів використали вольфрамовий конвертер. Товщина конвертера змінювалась від 0 до 6 мм. Для моделювання процесу опромінення нами була розроблена комп'ютерна програма мовою С++. Ця програма використовує бібліотеки класів Geant4, які базуються на методі Монте-Карло, і функціонує в багатопотоковому режимі. Для розрахунків була обрана модель "emstandard_opt3" складового модулю PhysicsList програми, як найбільш прийнятна у діапазоні енергій до 15 МеВ для процесів, що розглядаються. У роботі було визначено відносну кількість радіаційних пошкоджень, що припадають на один первинний електрон, та створений гамма-квант. Проведено порівняння результатів моделювання з експериментальними даними. На основі отриманих результатів зроблено висновки щодо основних механізмів, які призводять до розвалу молекул органічного барвника, а також запропоновано способи оптимізації експерименту для подальших досліджень.

Ключові слова: органічний барвник, електрон, гамма-квант, дозиметрія