COMPUTING AND MODELLING SYSTEMS

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THE COMPUTER MODEL EXPERIMENT FOR STUDYING MECHANISMS OF FORMATION OF RADIATION-STIMULATED DEFECTS IN MAGNESIUM-ALUMINUM SPINEL

S. Gokov[1], V. Horbach[2], S. Kochetov[1], L. Kolpakova[2], I. Lonina[1], O. Lyukhtan[1,2], E. Tsiats'ko[1]
[1]National Science Center "Kharkiv Institute of Physics and Technology", Kharkiv, Ukraine;
[2]V.N. Karazin Kharkiv National University, Kharkiv, Ukraine
E-mail: gokovsp@kipt.kharkov.ua

In the work in the Geant 4 software a computer model of the experiment on the study of the efficiency of formation of radiation-stimulated defects in magnesium aluminum spinel under irradiation with high-energy electrons and gamma quanta was developed. Under irradiation with electrons it was found that defects in the crystal lattice of spinel arise both under direct knocking out of atoms from the nodes and under photonuclear reactions. For energies of primary electrons of 25… 30 MeV the efficiency of formation of defects due to photonuclear reactions is several times higher compared to direct knocking out of atoms from the nodes of the crystal lattice. Under irradiation of samples with monoenergetic gamma quanta it was found that the efficiency of formation of defects is an order of magnitude higher than under irradiation with electrons of the same energy due to processes of photofission of nuclei.

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INTRODUCTION

Oxide ceramics and mono- and polycrystals of magnesium-aluminum spinel ($MgAl_2O_4$) are widely used in various sectors of the economy to create heat-resistant, catalytically active, optically transparent, electrical, chemically, corrosion- and radiation-resistant products. These properties ensure its successful use as portholes for spacecraft, windows for chemical reactors, sensors, membranes, structural materials for electrochemical fuel cells, catalysts and bases for them, electrical insulators, etc. [1–6]. To improve the reliability of nuclear reactors and solve environmental problems in nuclear power engineering, research is underway on the use of magnesium-aluminum spinel to create nuclear fuel with an inert matrix, targets for actinide transmutation, and matrices for isolating radioactive waste. For the successful use of magnesium-aluminum spinel for the above-mentioned tasks, additional research into its radiation resistance is necessary, in particular, research into the basic mechanisms of defect formation during irradiation with high-energy electrons and gamma quanta.

The main absorption bands corresponding to defects formed in spinel ceramic and crystals upon irradiation with ionizing radiation have been investigated in many works. Namely:

1. The absorption band at 2.8 eV, caused by hole centers on cation vacancies.
2. The absorption band at 3.8 eV, identified with hole centers on defects of the antistructure $(Mg^{2+}Al)^0$.
3. The absorption band at 4.15 eV, caused by electron centers on defects of the antistructure $(Al^{3+}Mg)^0$.
4. Anion vacancies with the capture of one electron form F+centers, the band is 4.75 eV [5].
5. Anion vacancies with the capture of two electrons form F-centers, the absorption band is 5.3 eV [6].

Thus, under high-energy ionizing irradiation, the process of formation of optical centers occurs through the creation of free charge carriers with subsequent capture by defects in the anionic or cationic sublattices of spinel crystals

When studying the absorption spectra, it was established that upon X-ray and gamma irradiation of spinel, the most likely formation of hole centers created mainly on defects on cation vacancies and defects in the antistructure. Upon irradiation with high-energy electrons or neutrons, radiation-induced centers are additionally created, and bands associated with F-types formed on anion vacancies appear in the absorption spectra. At the same time, it should be noted that there are significantly fewer studies of the mechanisms of the formation of defects in magnesium-aluminium spinel when it is irradiated with high-energy electrons than when exposed to X-rays and gamma radiation, so there is still no single presentation of the action of these mechanisms. For example, the dependence of the efficiency of the formation of defects on the density of the electron current, its energy, the probability of the occurrence of photonuclear reactions, and other things have not been studied much.

DESCRIPTION OF THE COMPUTER MODEL OF THE EXPERIMENT

The aim of this work is to study the efficiency of formation of radiation-stimulated defects in anionic and cationic sublattices of spinel crystals under irradiation with high-energy electrons and gamma quanta using a computer model of the experiment in the Geant 4 [7] software.

Modelling the displacement of atoms in a spinel ($MgAl_2O_4$) crystal lattice when exposed to high-energy particles such as electrons requires taking into account various physical processes associated with the interaction of charged particles with lattice atoms. Geant4 has standard physical processes for modelling ionization, scattering, and energy loss, but to account for atomic displacements, it is necessary to integrate a process that will track the amount of energy transferred to the atoms of the material and check whether this energy is sufficient to displace the atoms from their sites. For this purpose, a model describing particle-atom collisions **(Binary Collision Approximation)** was used.

First, the threshold displacement energy was determined for each atom in the spinel structure. This energy is the minimum energy required to knock an atom out of its crystal lattice site. For magnesium, aluminium, and oxygen, the values of the threshold displacement energy are different. In our model, the following values were chosen: Mg –30 eV, Al –40 eV, O –30 eV.

After determining the displacement threshold, the displaced atoms themselves, rather than the products of nuclear reactions, were determined from the set of knocked-out particles.

Natural magnesium consists of a mixture of 3 stable isotopes ^{24}Mg, ^{25}Mg, and ^{26}Mg with a molar concentration in the mixture of 78.6, 10.1, and 11.3%, respectively. Natural aluminium consists almost entirely of a single stable isotope ^{27}Al with a small content of ^{26}Al. Thus, the displaced atoms of ^{24}Mg, ^{25}Mg, ^{26}Mg, and ^{27}Al were determined as cation vacancies.

Oxygen has three stable isotopes: ^{16}O, ^{17}O, and ^{18}O, the average content of which is, respectively, 99.759, 0.037, and 0.204% of the total number of oxygen atoms on Earth. Thus, the displaced ^{16}O atoms were determined as anion vacancies. No displaced ^{17}O and ^{18}O atoms were observed during the work.

Formation of ^{12}C, ^{15}N, ^{8}Be nuclei associated with fission of ^{16}O nucleus was attributed to formation of anion vacancies due to photonuclear reactions.

The presence of a large number of ^{26}Al nuclei associated with fission of ^{27}Al nucleus with emission of a neutron was considered as cation vacancies with loss of aluminium atom from the crystal lattice site.

Formation of ^{23}Na, ^{20}Ne nuclei is associated with fission of ^{24}Mg nucleus and was also considered as cation vacancies with loss of magnesium atom from the crystal lattice site.

The most difficult to interpret was the presence of knocked-out ^{26}Mg nuclei, which could be formed both directly by knocking ^{26}Mg out of the crystal lattice site (cation vacancy with loss of Mg atom), and by fission of ^{27}Al with emission of a proton (cation vacancy with loss of Al atom). In this case, it was necessary to track the tracks of protons.

OPERATION WITH THE COMPUTER MODEL OF THE EXPERIMENT

A computer model of an experiment on studying the efficiency of formation of radiation-stimulated defects in magnesium aluminum spinel was developed in the Geant 4 programming environment. An electron beam from an accelerator was directed onto a $MgAl_2O_4$ sample measuring 1x1 cm and 1 mm thick and completely fell on it.

The developed model corresponded to the real experiment as much as possible. An algorithm for software detection of Mg, Al, O atoms knocked out of the crystal lattice nodes, which actually make up the studied defects, was developed. The sample volume itself was used as a sensitive detector, in which all secondary particles generated as a result of nuclear reactions and sample atoms that acquired kinetic energy due to interaction with primary electrons were registered. The screenshot of computer model of the experiment is shown in Fig. 1.

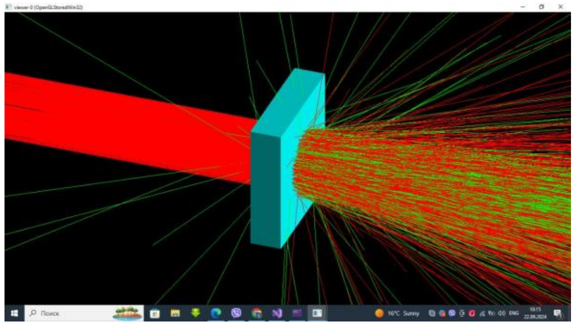

Fig. 1. The computer model of the experiment

Computer experiments were conducted on 10^8 primary electrons with energies of 5, 10, 15, 20, 2, and 30 MeV. At the primary particle energy of 5 MeV, nothing was observed. At an energy of 10 MeV, the program registered only sample atoms that acquired kinetic energy greater than 30 eV due to interaction with primary electrons. Starting with an energy of 12 MeV, we registered ^{23}Na, ^{12}C, ^{15}N, ^{26}Al, ^{23}Na which are products of photonuclear reactions. With an increase in the energy of primary electrons to 25 and 30 MeV, the number of both types of particles also increases.

Fig. 2 shows the efficiency of the formation of a cation vacancy associated with the knocking out or fission of the nucleus of an aluminum atom from a crystal lattice node.

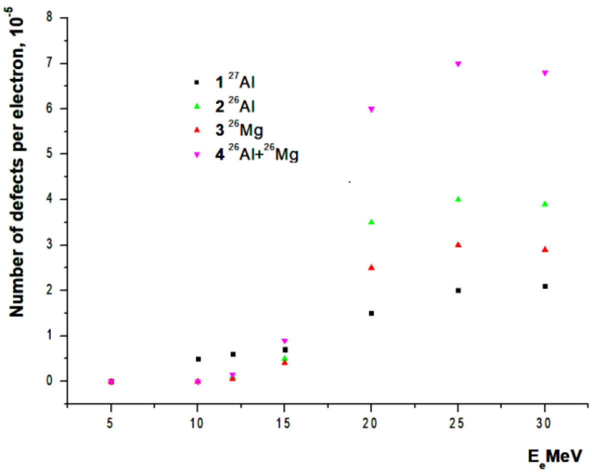

Fig. 2. The efficiency of the formation of a cation vacancies of an aluminum atom for electron beam energies 5, 10, 15, 20, 25, 30 MeV

Fig. 2 shows that the fission threshold of the ^{27}Al nucleus is in the region of the primary electron energy of 12 MeV. The number of nuclear fission events is significantly less than the number of atoms knocked out as a result of the interaction of primary electrons with the nucleus. Starting from the energy of 20 MeV, the number of cation vacancies formed as a result of photonuclear reactions increases significantly and at the energy of 25 MeV it exceeds the number of knocked out atoms by 6 times.

Fig. 3 shows the efficiency of the formation of a cation vacancy associated with the knocking out or fission of the nucleus of an magnesium atom from a crystal lattice node.

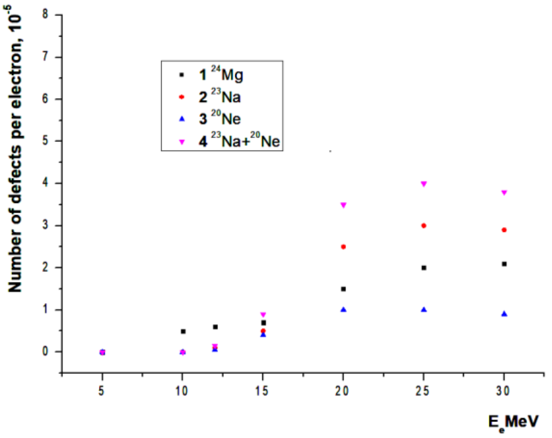

Fig. 3. The efficiency of the formation of a cation vacancies of an magnesium atom for electron beam energies 5, 10, 15, 20, 25, 30 MeV

It is also evident from the figure that the number of cation vacancies formed as a result of photonuclear reactions at an energy of 25 MeV is 4 times greater than the number of knocked-out atoms.

The dependence of the efficiency of vacancy formation on the target thickness was also investigated in the work. Fig. 4 shows the efficiency of formation of cation vacancies associated with knocking out or fission of the magnesium atom nucleus from a crystal lattice node with a change in target thickness from 1 to 10 mm for an electron energy of 25 MeV. All data were normalized to one incident electron and one thickness.

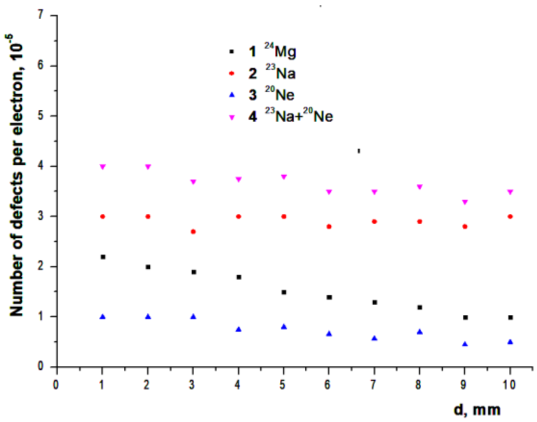

Fig. 4. The efficiency of formation of cation vacancies on the magnesium atom nucleus with a change in target thickness from 1 to 10 mm for an electron energy of 25 MeV

Fig. 4 shows that with an increase in the thickness of the target under study from 1 to 10 mm, the number of knocked-out particles per electron and one thickness decreases by 2 times. The number of nuclear reactions occurring in this range of thicknesses of the samples under study remains practically unchanged.

The work also included a study of the efficiency of defect formation in magnesium aluminum spinel when irradiated with monoenergetic gamma quanta with an energy of 10 to 40 MeV. Fig. 5 shows the efficiency of formation of cation vacancies associated with the knockout or fission of a magnesium atom nucleus from a crystal lattice site from the energy of incident gamma quanta.

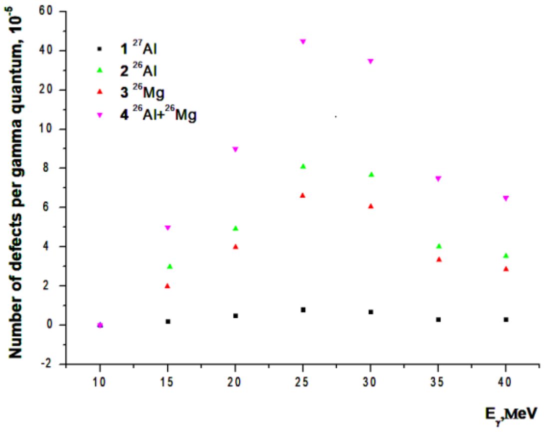

Fig. 5. The efficiency of the formation of a cation vacancies of an aluminum for gamma quanta beam energies 5, 10, 15, 20, 25, 30, 35, 40 MeV

Fig. 5 shows that the efficiency of defect formation due to fission reactions is an order of magnitude higher on gamma quanta than on electrons of the same energy. One can also see a maximum in the energy range of 25 MeV, which corresponds to the position of the giant dipole resonance for the elements included in the magnesium aluminum spinel. The presence of an insignificant number of magnesium atoms (no more than 5% of the total number of vacancies formed) is associated with the direct interaction of the products of photonuclear reactions with the spinel crystal lattice sites.

CONCLUSIONS

In the work in the Geant 4 software a computer model of the experiment on the study of the efficiency of formation of radiation-stimulated defects in magnesium aluminum spinel under irradiation with high-energy electrons and gamma quanta was developed. Under irradiation with electrons it was found that defects in the crystal lattice of spinel arise both during direct knocking out of atoms from the nodes and during photonuclear reactions due to the occurrence of bremsstrahlung gamma quanta during the interaction of high-energy electrons with a sample of magnesium aluminum spinel. In this case, the threshold of photonuclear reactions is in the range of 12…13 MeV for various chemical elements included in the composition of magnesium aluminum spinel. In the work it was found that for primary electron energies of 25…30 MeV the efficiency of defect formation due to photonuclear reactions is several times higher compared to the direct knocking out of atoms from the nodes of the crystal lattice. When irradiating samples with monoenergetic gamma quanta, it was found that the efficiency of defect formation was an order of magnitude higher than when irradiating with electrons of the same energy due to photofission processes of nuclei.

A maximum of defect formation was also observed in the energy region of 25 MeV, which corresponds to the position of the giant dipole resonance for the elements that make up magnesium aluminum spinel. In this case, an insignificant number of target atoms (no more than 5% of the total number of formed vacancies) were knocked out by direct interaction of photonuclear reaction products with the nodes of the spinel crystal lattice.

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КОМП'ЮТЕРНИЙ МОДЕЛЬНИЙ ЕКСПЕРИМЕНТ ДЛЯ ДОСЛІДЖЕННЯ МЕХАНІЗМІВ УТВОРЕННЯ РАДІАЦІЙНО-СТИМУЛЬОВАНИХ ДЕФЕКТІВ У МАГНІЄВО-АЛЮМІНІЄВІЙ ШПІНЕЛІ

С. Гоков, В. Горбач, С. Кочетов, Л. Колпакова, І. Лоніна, О. Люхтан, Є. Цяцько

У програмному середовищу Geant 4 була розроблена комп'ютерна модель експерименту з дослідження ефективності утворення радіаційно-стимульованих дефектів магнієво-алюмінієвої шпінелі при опроміненні високоенергетичними електронами та гамма-квантами. При опроміненні електронами було встановлено, що дефекти в кристалічній решітці шпінелі виникають як при безпосередньому вибиванні атомів із вузлів, так і при протіканні фотоядерних реакцій. Для енергій первинних електронів 25…30 МеВ ефективність утворення дефектів внаслідок протікання фотоядерних реакцій у кілька разів вища порівняно з безпосереднім вибиванням атомів із вузлів кристалічної решітки. При опроміненні зразків моноенергетичними гамма-квантами було встановлено, що ефективність утворення дефектів на порядок вища, ніж при опроміненні електронами тієї ж енергії за рахунок фотоподілу ядер.