THE STUDY OF MECHANISMS RADIATION-STIMULATED DEFECTS FORMATION IN THE MAGNESIUM-ALUMINUM SPINEL UNDER ELECTRON IRRADIATION

S. Gokov[1], S. Kochetov[1], A. Kozlovskyi[3], O. Lyukhtan[1,2], S. Nizhankovskyi[3], Yu. Siryk[3], 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;
[3]Institute for Single Crystals, Kharkiv, Ukraine

The paper investigated the effectiveness of the formation of radiation-stimulated defects in magnesium-aluminum spinel upon irradiation with high-energy electrons. Initially, a computer model of the experiment was developed in the Geant 4 software environment. The simulation results showed that defects in the crystal lattice of spinel occur both when atoms are directly knocked out of nodes and when photonuclear reactions occur when irradiated with electrons with energy above the photofission threshold. Single-crystal spinel samples were irradiated with primary electron energies of 10, 15, 20, and 25 MeV in the atmosphere, with a fluence of $2 \cdot 10^{16}$ cm^{-2}, on the LUE-30 linear electron accelerator of the NSC KIPT. The obtained experimental results, as well as the modeling, showed that photonuclear reactions play a significant role in the formation of defects in spinel crystals under electron irradiation.

PACS: 61.72.Cc, 61.80.Hg, 78.20.Ci, 87.80.+s, 87.90.+y, 07.05.Tp, 78.70.–g

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.

The aim of this work is to study the efficiency of formation of radiation-stimulated defects in anion and cation sublattices of spinel crystals under irradiation with high-energy electrons. The work was carried out in two stages. At the first stage, a computer model as close as possible to the experiment was created in the Geant 4 programming environment [7]. At the second stage, magnesium aluminum spinel crystal samples were irradiated at the LUE-30 linear electron accelerator of the NSC KIPT**.**

THE COMPUTER MODEL OF THE EXPERIMENT

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 1 x 1 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.

Computer experiments were conducted on 10^8 primary electrons with energies of 5, 10, 15, 20, 25 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 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. 1 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.

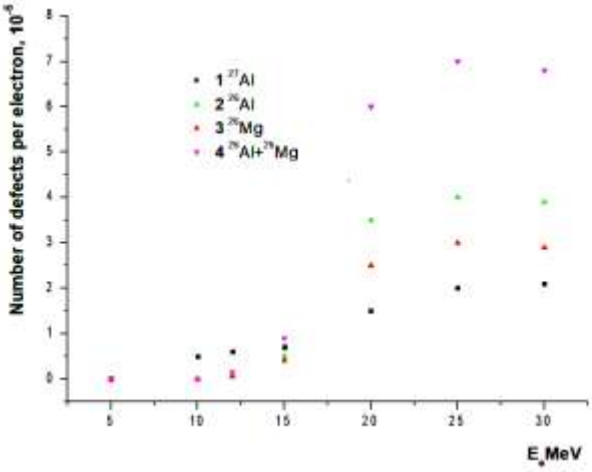

Fig. 1. The efficiency of the formation of a cation vacancies of an aluminum atom for electron beam energies 5, 10, 15, 20, 25, 30 MeV

Fig. 1 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. 2 shows the efficiency of the formation of a cation vacancy associated with the knocking out or fission of the nucleus of the magnesium atom from a crystal lattice node.

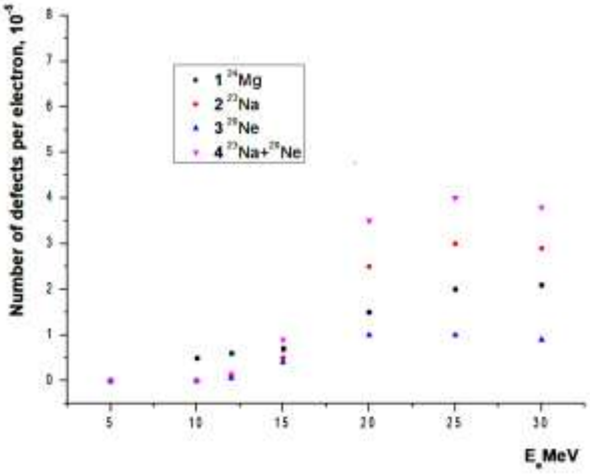

Fig. 2. The efficiency of the formation of the cation vacancies of the magnesium atom for electron beam energies 5, 10, 15, 20, 25, 30 MeV

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.

EXPERIMENTAL INVESTIGATION OF MAGNESIUM-ALUMINUM SPINELS

The paper presents an experimental study of magnesium-aluminum spinel samples. The aim of the work was to investigate the dependence of the efficiency of defect formation in magnesium-aluminum spinel at low fluence on the electron beam energy. Experiments were conducted with primary electron energies of 10, 15, 20 and 25 MeV. The total fluence did not exceed $2x10^{16}$ e/cm^2. Two samples were used for the study. They were grown and prepared at the Institute for Single Crystals. They are round, 10 mm in diameter and 2 mm thick. All samples are polished on both sides.

Fig. 3. The photo of the experiment on spinel irradiation

The samples were irradiated at the output of the linear electron accelerator LUE-30 with a small current. The current density did not exceed 2 μA/cm^2. For good cooling, the samples were placed on thermal paste on an aluminum radiator and blown with an air flow. A photograph of the experiment is shown in Fig. 3.

The samples before and after irradiation are presented in Fig. 4.

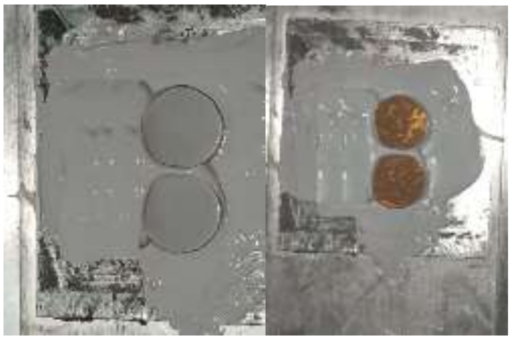

Fig. 4. The spinel samples before irradiation (left) after irradiation (right)

After irradiation, the transmission and absorption spectra of the samples were studied using a spectrophotometer OPTIZEN 3220 UV.

When processing the experimental data, it was found that for targets 1 and 2, the absorption spectra for one energy almost completely coincide. This indicates a uniform flow of electrons at the location of the samples.

The optical absorption spectra of sample №2 induced by electron irradiation with energy of 10, 15, 20, 25 MeV up to a fluence of $2\cdot10^{16}$ e/cm^2 are shown in Fig. 5.

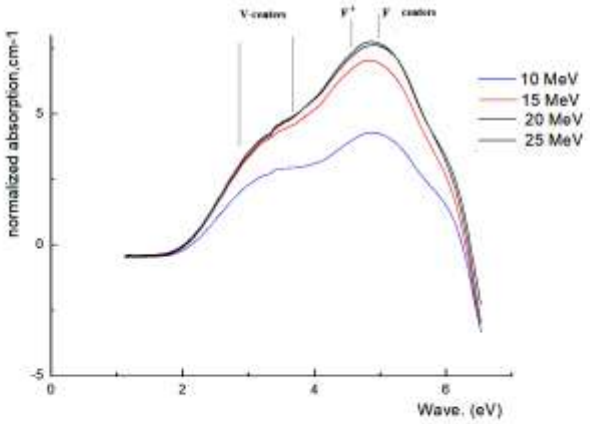

Fig. 5. The optical absorption spectra of sample № 2 induced by electron irradiation with energy of 10, 15, 20, 25 MeV

It can be seen from Fig. 5 that peaks with maxima at 4.75, 5.3 eV and a complex band of optical V-centers on cation vacancies increase rapidly when the energy of the electron beam increases from 10 to 15 MeV. Further, this growth continues up to an electron beam energy of 20 MeV and enters saturation. Such an increase in the number of defects is probably related to the course of photonuclear reactions, which was also assumed in the computer model.

Thus, photonuclear reactions play a significant role in the creation of defects in magnesium aluminum spinel.

CONCLUSIONS

In the work in the Geant 4 environment, a computer model was developed and tested to study the processes of formation of radiation-stimulated defects in magnesium-aluminum spinel.

Using the model, the processes of defect creation in spinel were simulated depending on the electron beam energy in the range of 5…40 MeV.

A number of experiments were conducted on the LUE-30 linear electron accelerator to study the efficiency of defect formation from the electron beam energy for energies of 10, 15, 20, 25 MeV.

The obtained experimental results, as well as the modeling, showed that photonuclear reactions play a significant role in the formation of defects in spinel crystals under electron irradiation.

REFERENCES

1. S.J. Zinkle, H. Matzke, V.A. Skuratov. Microstructure of swift heavy ionirradiated $MgAl_2O_4$ spinel// *Mat. Res. Soc. Symp. Proc.* 1999, v. 540, p. 299-304.

2. M. Ishimaru, Y. Hirotsu, I.V. Afanasyev-Charkin, K.E. Sickafus. Atomistic structures of metastable and amorphous phases in ion-irradiated magnesium aluminate spinel // *J. Phys, Condens. Matter.* 2002, v. 14, p. 1237-1247.

3. L. Thomé, A. Gentils, J. Jagielski, F. Garrido, T. Thomé. Radiation stability of ceramics: Test cases of zirconia and spinel // *Vacuum.* 2007, v. 81, p. 1264-1270.

4. F.A. Garner, G.W. Hollenberg, F.D. Hobbs, J.L. Ryan, Z. Li, C.A. Black, R.C. Bradt. Dimensional stability, optical and elastic properties of $MgAl_2O_4$ spinel irradiated in FFTF to very high exposures // *J. Nucl. Mater.* 1994, v. 212-215, p. 1087-1090.

5. G.S. White, K.H. Lee, J.H. Crawford, Jr. Effect of γ-irradiation upon the optical behavior of spinel // *Phys. Status Solidi.* 1977, v. A 42, p. K137-141.

6. G.P. Summers, G.S. White, K.H. Lee, J.H. Crawford, Jr. Radiation damage in $MgAl_2O_4$ // *Phys. Rev. B.* 1980, v. 21, p. 2578-2584.

7. geant4-userdoc.web.cern.ch/UsersGuides/ForApplicationDeveloper/BackupVersions/V10.6c/fo/BookForApplicationDevelopers.pdf