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EFFECT OF ELECTRON IRRADIATION CONDITIONS ON THE EFFICIENCY OF DEFECT FORMATION IN $MgAl_2O_4$ SPINEL

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The efficiency of defect formation in spinel ceramics of magnesium aluminate ($MgAl_2O_4$) under electron irradiation has been determined. A strong effect of cooling on the concentration of residual defects was revealed. When using electrons with energies above the giant dipole resonance for Mg, Al, O and cooling the sample with an air-blown aluminum radiator, a total increase in the efficiency of F-center formation by a factor of 4.3 was obtained.

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INTRODUCTION

Magnesium aluminum spinel crystals and ceramics are known for their exceptional resistance to neutron and ion irradiation. Irradiation by fast electrons is used to simulate the radiation effect on thermonuclear devices. The cross-sections for the production of defects by relativistic electrons in spinel reach a threshold at electron energies above 10 MeV and do not exceed 10 barns for oxygen and 35 barns for Mg and Al [1]. Therefore, to achieve concentrations of radiation defects corresponding to conditions similar to ITER, a very long exposure is required. At electron energy of 2.5 MeV and beam current density of 5.9 $\mu A/cm^2$, F-centers concentration of $2.5 \cdot 10^{18}$ cm^{-3} was obtained for irradiation during 55 hours and this corresponds only to $2.5 \cdot 10^{-5}$ dpa.

An increase in the energy of electrons from 2.5 to 50 MeV leads to an increase in the efficiency of creating primarily shifted atoms only twice [2]. And a significant increase in beam current is limited by the possibility of removing heat from the samples.

But at electron energies over 20…25 MeV, the channel of photonuclear reactions on the nuclei of crystal-forming atoms is additionally activated. Despite the very small cross-section of the photonuclear reaction, each recoil nucleus with energy of up to 0.5 MeV produces a large-sized defect cluster, which significantly increases the efficiency of creating stable defects [3].

1. EXPERIMENTAL DETAILS

Spinel samples were irradiated in three different conditions using two accelerators. Two electron beam irradiation schemes were developed at the 30 MeV Electron Linac irradiation facility of the Kharkiv Institute of Physics and Technology of the National Academy of Sciences of Ukraine. The pulse repetition rate and duration were 50 Hz and about 2 μs, respectively.

In the first irradiation session, the electron energy was 16 MeV, the average beam current density was 2.5 $\mu A/cm^2$, and different fluencies were available by variation the irradiation time. Ceramic samples were placed on 2 mm glass substrate with intensive air cooling. The glass substrate was used to determine the electron beam dimensions and electron beam print coordinate location.

The second series of irradiation experiments were carried out with electron energy of 25 MeV, electron beam current density of 0.65 $\mu A/cm^2$. Spinel samples had contact with a 2 mm air-cooled aluminum plate by KPT-19 thermal paste. The sketch of the experimental setup is shown in Fig. 1. Two similar exposures were developed at this energy with 100 μm tungsten foil and without it. It was placed at a 5 mm distance in front of the sample.

The third irradiation experiment was carried out on the M-30 microtron of the Institute of Electronic Physics of the National Academy of Sciences, Uzhgorod. The electron energy was 12.5 MeV, and the electron beam density equaled 4 $\mu A/cm^2$. The pulse repetition rate was 1 kHz, and the pulse duration was 0.3 ns.

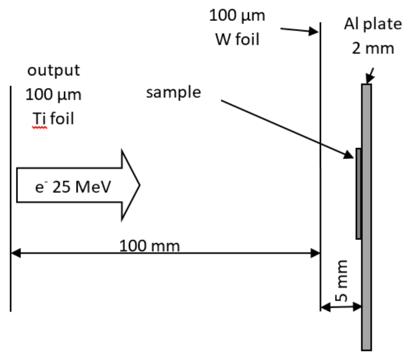

Fig. 1. Sample placement during 25 MeV electron irradiation

Stoichiometric spinel ceramics were obtained by the usual method of hot pressing spinel powder containing 1 % lithium fluoride to obtain an optically transparent material. Sections measuring 12x7x0.7 mm were cut and also polished for optical processing.

The concentration of radiation defects in spinel samples was measured by the optical absorption method and calculated according to Smakula equation [4]. Absorption spectra measurement was provided with a single beam SF-46 spectrophotometer.

Some samples were subjected to isochronous annealing. The temperature step was chosen to be 50°C, and the annealing time was 30 s.

2. RESULTS AND DISCUSSION

Optical absorption spectra of ceramics induced by irradiation with 16 MeV electrons to fluences in the range from $4.2\cdot10^{16}$ to $3.3\cdot10^{17}$ e/cm^2 are shown in Fig. 2. As can be seen, the peak with a maximum at 5.3 eV gradually increases with increasing fluence, while the complex band of optical V-centers on cationic vacancies at 3…4 eV remains the same intensity.

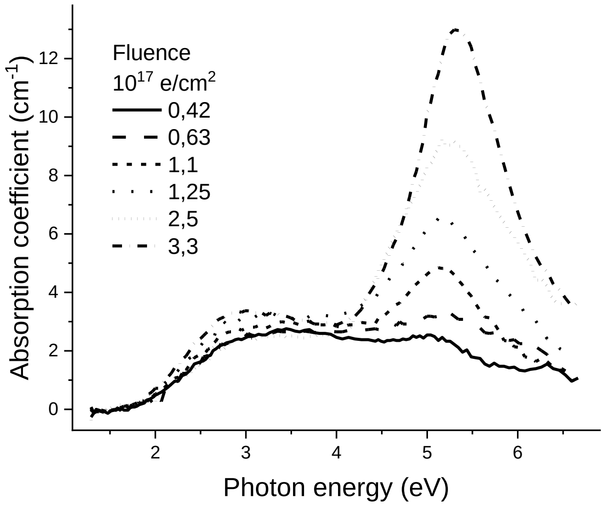

Fig. 2. Optical absorption induced by 16 MeV electron irradiation to different fluence in spinel ceramics

Because of isochronal annealing of radiation-induced absorbance at the first 50 degrees step mainly vanishing absorption bands of V-centers, we have concluded that the temperature of samples during irradiation didn't exceed 50°C (Fig. 3).

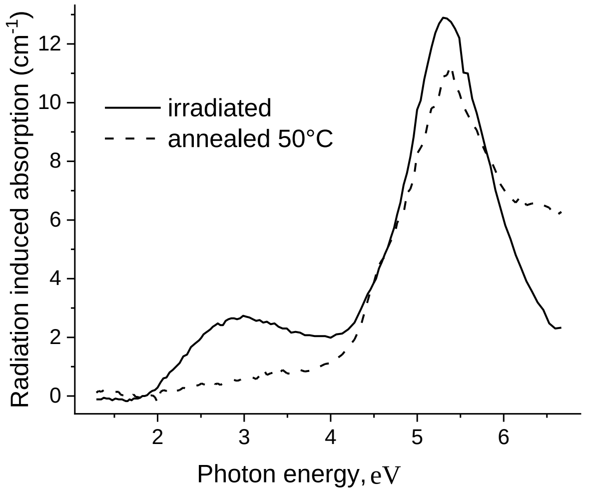

Fig. 3. Optical absorption induced by 16 MeV electron irradiation to fluence $3.3\cdot10^{17}$ e/cm^2 in spinel ceramics

Since the direct measurement of the temperature of the sample is associated with certain difficulties at pulsed accelerator mode, a theoretical analysis of the thermal balance of the samples during irradiation was carried out. Assuming electron energy losses for ionization in spinel equal to 0.7 MeV/mm [5] we obtain that the temperature jump during the electron pulse is approximately 0.04 K/μA. That is, at current density of up to 10…20 μA/cm^2, the temperature of the sample is quite constant. The energy balance equation gives the stationary temperature of the sample:

$$T_{\max} = T_o + j\frac{dE}{dx} l \cdot R, \qquad (1)$$

where T_0 is the room temperature, j is the beam current density, dE/dx is ionization loss, l is the sample thickness, and R is the thermal resistance of the sample-atmosphere junction. For spinel samples placed on a glass surface, R-value reaches 200 K/W and the temperature could rise to 200°C. By using a heat sink thermal resistance could be sufficiently decreased so the temperature of the sample too. To test this assumption, irradiation was carried out at electron energy of 12.5 MeV and fluence of $6\cdot10^{16}$ e/cm^2 under conditions as shown in Fig. 1, but without W foil. Irradiation induced optical absorption together with the mainly the same fluence of $6.3\cdot10^{16}$ e/cm^2 at energy 16 MeV are shown in Fig. 4. Surface square of the aluminum plate equaled to 200 cm^2.

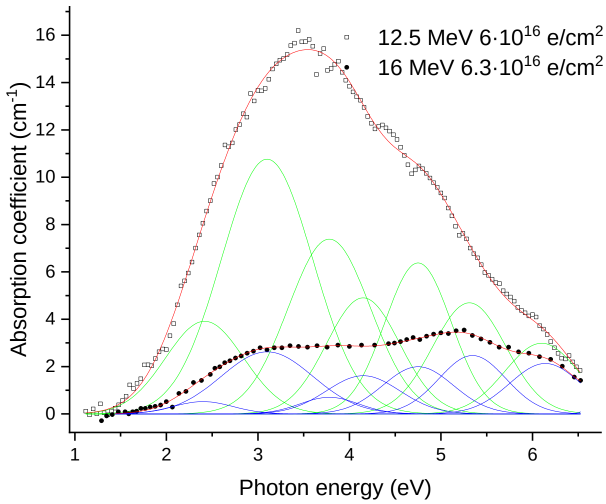

Fig. 4. Radiation-induced optical absorption of spinel ceramics irradiated by electrons by using different cooling setups

It is easy to see that, despite the lower electron energy, the efficiency of forming optical centers is much higher when using a radiator. Fitting the optical spectra with *Gaussian peaks* corresponding to [6] shows that the concentration of *F*-centers (5.3 eV) is almost the same in both cases, while the other centers are significantly suppressed for the sample irradiated on glass. Such a more noticeable difference in the results of exposure indicates the importance of using a radiator during exposure unless liquid nitrogen or helium is used to cool the sample.

According to the scheme of the experiment shown in Fig. 1, two irradiation experiments were carried out with electron energy of 25 MeV and relatively low fluence of $1.9\cdot10^{16}$ e/cm^2. Radiation-induced absorption spectra of samples irradiated with a direct electron beam (lower open dot) and through tungsten foil with 100 μm thickness of (upper filled dot) are shown in Fig. 5.

Conditions of 25 MeV electron irradiation and $1.9\cdot10^{16}$ e/cm^2 fluence provide intermediate effect between 12.5 MeV with an aluminum radiator and irradiation on glass at 16 MeV, but with a fluence of $6\cdot10^{16}$ e/cm^2. The introduction of 100 μm tungsten converter increases the efficiency of F-center production by 1.6 times and very slightly for V-centers.

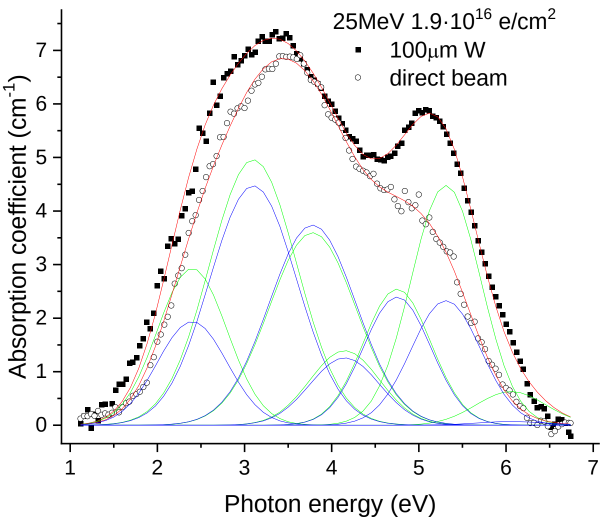

Fig. 5. Radiation-induced optical absorption of spinel ceramics irradiated with 25 MeV electrons, fluence $1.9 \cdot 10^{16}$ e/cm^2, with (black squares) and without tungsten foil

Since 100 μm is small compared to the tungsten radiation length, most of the electrons passing through the foil have essentially the same effect on the sample as a direct electron beam. The addition defects were produced by high-energy γ-quanta through the γ-n reaction. But it should be noted that the titanium foil of the output window of the accelerator is also a converter. Although it compared to tungsten is less effective.

The maxima of the giant dipole resonance for spinel components are in the range of 18…24 MeV with a peak cross-section of 10…28 mb [7]. This is three orders of magnitude smaller than the cross sections for elastic scattering of electrons, but significantly increases the total concentration of radiation defects. Concentration on radiation-induced optical centers in spinel ceramics was calculated via the Smakula equation and is summarized in Table. The atomic oscillator strength for F-centers [8] was assumed equal to 1 and 0.1 for V-centers [9].

Radiation-induced optical center concentration in spinel ceramics after electron irradiation at different conditions

Electron Energy, MeV	Fluence, 10^{16} e/cm^2	Irradiation conditions	F-centers, 10^{16} cm^{-3}	V-centers, 10^{16} cm^{-3}
12.5	3	on Al	2.1	72
12.5	6	on Al	2.5	75
16	6.3	on glass	1.2	18.4
16	33	on glass	5.6	25
25	1.9	on Al + W	1.9	30
25	1.9	on Al	1.2	29

V-center concentration comparison shows that it depends more on the irradiation conditions than on the electron fluence: with the same fluence of $(6…6.3) \cdot 10^{16}$ e/cm^2, the concentrations differ by a factor of 4.3 and almost the same concentrations are realized with fluence ratio of 17. Such behavior of hole centers corresponds to a strong suppression of the creation of stable cation vacancies (and hole centers) in the spinel crystal due to cation inversion and radiation-induced annihilation of Frenkel pairs [10]. The highest concentration of V-centers obtained in this work is about 10^{18} cm^3, which corresponds to the thermodynamic concentration of vacancies that can be formed at the sintering temperature of ceramics of 1380°C with a vacancy formation energy of 1.6 eV [11]. The practically absent dependence of the concentration of hole centers on the fluence indicates that changing the irradiation conditions allows for an increase in the part of optically active cation vacancies that were formed during growth.

F-center concentration for all spectra from Fig. 2 (not just two from Table) along with other samples depending on the electron fluencies is shown in Fig. 6. The influence of the converter at electron energy of 25 MeV was taken into account by manually increasing the fluence in proportion to F-center concentration.

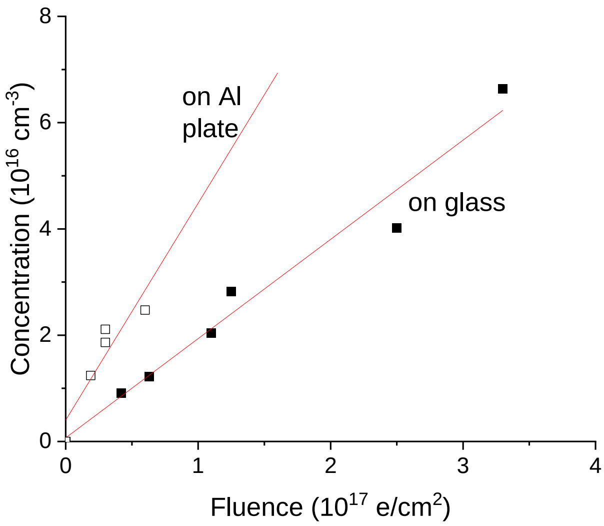

Fig. 6. Dependence of F-center concentration on electron fluence for two cooling setups

Straight lines in Fig. 6 are independent linear fitting of F-center concentration obtained with glass substrate and aluminum heat sink during irradiation. The effectiveness of anion vacancy creation at elevated temperatures is 0.18 vacancy per electron. For the aluminum cooling system effectiveness rises to 0.4.

CONCLUSIONS

The use of an aluminum plate as a heat sink makes it possible to increase the efficiency of F-center formation by a factor of 2. And this is in addition to a 4-fold increase in the concentration of V-centers.

Irradiation with electrons through thin e-γ converter with the electron energy above dipole resonance additionally increase the efficiency of F-center production by 60 %. The total increase in the efficiency of F-center formation due to cooling and the use of a gamma converter reaches 4.3 times and can possibly be further improved by optimizing the thickness of the converter.

Unlike F-centers, V-centers do not exhibit a significant dependence on electron fluence under the experimental conditions used. But such a condition strongly determines the residual concentration of V-centers: it increases with decreasing temperature during irradiation.

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ВПЛИВ УМОВ ЕЛЕКТРОННОГО ОПРОМІНЕННЯ НА ЕФЕКТИВНІСТЬ ДЕФЕКТОУТВОРЕННЯ В ШПІНЕЛІ $MgAl_2O_4$

Ю.Г. Казарінов, О.М. Поп, І.Г. Мегела, А.І. Попов

Визначено ефективність дефектоутворення при електронному опроміненні в кераміці магній-алюмінієвої шпінелі ($MgAl_2O_4$). Виявлено сильний вплив охолодження на концентрацію залишкових дефектів. При використанні електронів з енергією вище гігантського дипольного резонансу для Mg, Al, O та охолодженні зразка алюмінієвим радіатором з повітряним обдувом отримано сумарне підвищення ефективності формування F-центрів у 4,3 рази.