EFFICIENCY OF DEFECTS FORMATION IN SPINEL UNDER HIGH ENERGY ELECTRON AND GAMMA BEAM IRRADIATION

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There were provided the comparative investigations of the efficiency of optical absorption centers formation in magnesium aluminates spinel (single crystals and ceramics) at the influence of high energy gamma or electron beams. It was revealed that at gamma irradiation in the single crystals and ceramics the most probably the hole centers are formed which were created primarily at growth defects: cationic vacancies and anti-site defects. After electron irradiation of different fluences in absorption spectra there were observed bands related to hole centers (defects in the cationic sub-lattice) which practically does not change, while the concentration of electron centers (defects in the anion sub-lattice) grows proportionally to the electron fluence.

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1. INTRODUCTION

Magnesium aluminates spinel, $MgAl_2O_4$, (hereafter termed "spinel") is highly radiation resistant material and is prospective for use in nuclear reactors as fuel forms, as inert matrix for transmutation of actinides, in fusion reactors as insulators for magnetic coils and windows for radiofrequency plasma heating [1, 2]. In all indicated cases this material w frequently will undergo to irradiation of different nature and intensity. Because single crystals are restricted in size and production is very expensive the development of ceramic technology of this material is now in progress [3, 4]. Therefore, the comparison of influence of irradiation on the properties of single crystals and ceramics is issue of the day. One of the most successful processes for producing optical quality spinel ceramics is hot-pressing technique of spinel powder doped with LiF [5, 6]. The variations of processing condition were applied to change of microstructure, porosity, optical and mechanical properties of spinel ceramics. Thus initial properties of ceramics and single crystals, basically degree of crystalline perfection, may influence on the sensitivity of this material to different types of irradiation. Under high energy ionizing irradiation such as gamma-rays or fast electrons the process of optical center formation goes through creation of free charge carriers with subsequent trapping by growth or radiation induced defects and impurity ions. In this paper we describe the results of comparative investigations of radiation induced defects in spinel single crystals and ceramics after irradiation with high energy electrons and gamma quanta.

2. EXPERIMENTAL PROCEDURE

Irradiation of spinel samples with electron or gamma beam was provided at linear accelerator $LUE-300$ with nominal electron energy of $30\,MeV$. There was used deflected output which allows to decrease the energy spread of electron beam and contribution of bremsstrahlung gamma-rays at irradiation. The energy of electron was $16\,MeV$, the beam current about 2.5 and $10\,\mu A/cm^2$, and different fluences was accessed by variation of irradiation time. The irradiation with gamma beam was provided at strait output to increase the irradiation dose. Gamma-rays were generated by conversion of electrons with energy of $7\,MeV$ in tantalum target with thickness of $2.0\,mm$. Using deflecting magnet the gamma component was separated from electrons. Gamma-beam was formed using collimator and was directed to samples. Electron fluence on conversion target was 3.4×10^{17} electrons, which corresponds to 10^{16} $gamma\,quant/cm^2$ on samples under investigation. Stoichiometric spinel crystals were grown by Verneuil methods. From grown boule the slices of $12\times10\times0.7\,mm$ were cut and polished to optical finish. Stoichiometric spinel ceramics were obtained by ordinary hot-pressing technique of spinel powder containing $1\,\%$ of lithium fluoride to get optically transparent material. Slices of $12\times7\times0.7\,mm$ were cut and also polished to optical finish.

3. RESULTS AND DISCUSSION

The absorption spectra of pristine crystals have no definite bands indicating the

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absence of impurities. In the gamma-ray-irradiated samples the absorption spectra contains several overlapping bands which form maxima at approximately 3.1 and $5.0\,eV$ (Fig.1).

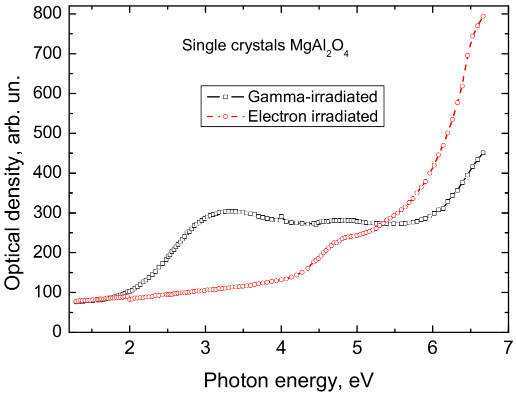

Fig.1. *Optical absorption spectra of gamma- and electron-irradiated spinel single crystals*

Previous investigations of radiation induced optical centers gave a possibility to identify the absorption bands with corresponding lattice defects: $5.3\,eV$ absorption band is related to F-centers, i.e. anion vacancies captured two electrons [7, 8]; $4.75\,eV$ absorption related to F^+ - centers, also cation vacancies but captured one electron [9, 10]; $4.2\,eV$ absorption band was tentatively identified with electron centers formed at defects of positively charged anti-site defects. Absorption of photons of energy lower $4\,eV$ contains many overlapping bands which were identified with different types of hole centers, i.e. cationic vacancies and negatively charged anti-site defects captured one or more holes [11, 12]. Therefore we conclude that under gamma irradiation both hole and electron centers of different concentration are formed. It should be noted that absorption in high energy photon range very often caused by uncontrolled impurities which change under irradiation charging states.

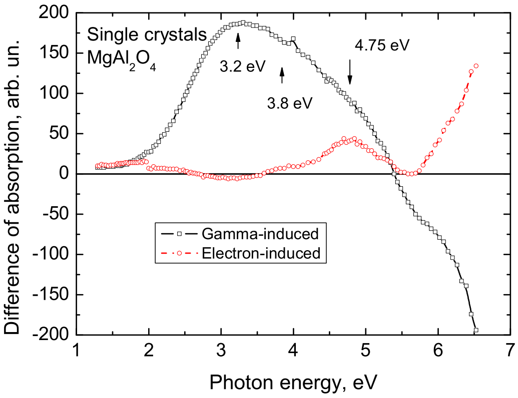

Fig.2. *Radiation induced optical absorption spectra in gamma- and electron-irradiated spinel single crystals*

Electron irradiation to fluence of $3.0 \times 10^{16} el/cm^2$ at the current density of $10\,\mu A/cm^2$ leads to formation only one weak absorption band at $4.75\,eV$. To deconvolute the spectra into specific bands we subtract from spectra of irradiated sample the initial spectra. Such difference absorption spectra are shown in Fig.2. Evidently we can distinguish main band at 3.2 and $4.75\,eV$ with some contribution of bands at $3.8\,eV$ (as a peak) and $5.3\,eV$ (as a hole) [13]. Therefore, we may conclude that under gamma irradiation the main process of optical centers formation is charge exchange between cationic and anionic vacancies. The irradiation with high energy electrons at high flux the temperature of sample was raise up to $200^\circ C$, which leads to annealing of hole centers, also the radiation created anionic vacancies capture electrons forming F^+ centers, which are stable up to $200^\circ C$ [14]. Absorption spectra of gamma irradiated spinel ceramics very similar to that of single crystals, which are shown in Fig.3. Absorption spectra of electron irradiated ceramics at low electron flux $2.5\,\mu A/cm^2$ to different fuences demonstrate the formation of two wide spectral regions: in the visible range of photon energy $2...4\,eV$ which does not depend on fluence, and UV-range of photon energy $4...6\,eV$ growing with increase of fluence. By subtracting of initial spectra we obtained radiation induced optical spectra, which are shown in Fig.4. By deconvolution of these spectra into Gaussians we obtained bands at 2.8, 3.2 and $3.8\,eV$ which were identified with hole centers at cationic defects, including empty cationic vacancy, complex defects of lithium ions in cation vacancy, and anti-site defects $(Mg^{2+}_{Al})^0$, respectively. In the second spectral region the bands at 4.15, 4.75 and $5.3\,eV$ are found which were identified with electron centers at anion defects, related to anti-site defects $(Al^3 +_{Mg})^0$, anion vacancies captured one electron (F+-centers) or two electrons (F-centers), respectively. In the indicated range of fluences the concentration of hole centers practically does not change (defects in cationic sub-lattice), at the same time the concentration of electron centers

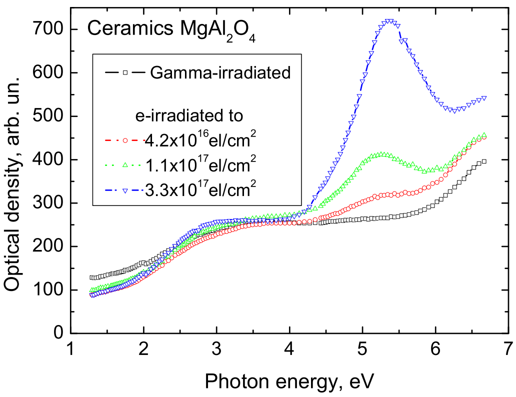

Fig.3. *Optical absorption spectra of gamma- and electron-irradiated spinel ceramics*

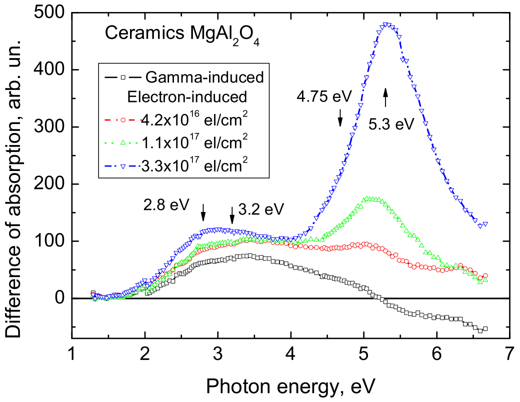

Fig.4. *Radiation induced optical absorption spectra in gamma- and electron-irradiated spinel ceramics*

(defects in anion sub-lattice) growths proportionally to electron fluence. Using semi empirical Smakula formula we calculated the maximal concentration of F-type centers which reaches $1.1 \times 10^{17}\,defects/cm^3$. I.e., the efficiency of creation of stable defects at irradiation with $16\,MeV$ electrons equals $0.33\,defects/electron$.

4. CONCLUSION

There were provided the comparative investigations of the efficiency of optical absorption centers formation in magnesium aluminates spinel (single crystals and ceramics) at the action of high energy gamma or electron beams. It was revealed that at gamma irradiation of single crystals the most probably the hole centers are formed which were created primarily at growth defects: cationic vacancies and anti-site defects. At the irradiation with electrons at beam current density of $10\,\mu A/cm^2$ the temperature of sample was raised which leads to partly thermal annealing of unstable radiation-induced centers and absorption spectra demonstrate the weak bands related of F-type centers which are formed at radiation induced anionic vacancies. After gamma irradiation of spinel ceramics also mainly hole centers are formed at initial defects. After electron irradiation of ceramics at beam current density of $2.5\,\mu A/cm^2$ in absorption spectra there were observed bands related to hole and electron centers. At the investigation of the dose dependences of the efficiency of defects formation it was found that the concentration of hole centers (defects in the cationic sub-lattice) practically does not change, while the concentration of electron centers (defects in the anion sub-lattice) grows proportionally to the fluence of irradiation. There was defined the efficiency of defect formation in anionic vacancies in complex oxide of magnesium aluminates spinel at the irradiation with $16\,MeV$ electrons.

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ЭФФЕКТИВНОСТЬ ОБРАЗОВАНИЯ ДЕФЕКТОВ В ШПИНЕЛИ ПОД ВОЗДЕЙСТВИЕМ ВЫСОКОЭНЕРГЕТИЧНЫХ ЭЛЕКТРОНОВ И ГАММА-КВАНТОВ

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Проведены сравнительные исследования эффективности образования оптических центров поглощения в магний-алюминиевой шпинели (монокристаллов и керамики) при воздействии высокоэнергетических гамма-квантов и электронов. Установлено, что гамма облучение монокристаллов и керамики приводит преимущественно к образованию дырочных центров, которые формируются на ростовых дефектах: катионных вакансиях и дефектах антиструктуры. После электронного облучения до различных флюенсов в спектрах поглощения наблюдаются полосы, обусловленные дырочными центрами (дефекты в катионной подрешетке), концентрация которых практически не изменилась, в то время как концентрация электронных центров (дефектов в анионной подрешетке) растет пропорционально флюенсу электронов.

ЕФЕКТИВНІСТЬ УТВОРЕННЯ ДЕФЕКТІВ В ШПІНЕЛІ ПІД ДІЄЮ ВИСОКОЕНЕРГЕТИЧНИХ ЕЛЕКТРОНІВ ТА ГАМА-КВАНТІВ

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Виконано порівняльні дослідження ефективності створення оптичних центрів поглинання в магній-алюмінієвій шпінелі (монокристалів та кераміки) під дією високоенергетичних гама-квантів та електронів. Показано, що гама-опромінення монокристалів та кераміки призводить переважно до створення діркових центрів, які формуються на ростових дефектах: катіонних вакансіях та дефектах антиструктури. Після електронного опромінення різними флюенсами в спектрах поглинання спостерігаються смуги, зумовлені дірковими центрами (дефекти в катіонній підгратці), концентрація яких практично не змінилась, в той час як концентрація електронних центрів (дефекти в аніонній підгратці) росте пропорційно флюєнсу електронів.