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EFFECT OF ELECTRIC FIELD ON TRANSPORT OF RADIATION DEFECTS IN MgO

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After irradiation by neutrons the absorption spectra of MgO consists of 250, 357 and 570 nm bands. The accumulation of radiation defects in MgO doped with nickel differs from that in pure MgO. The intensity of the 570 nm absorption band is considerable smaller in MgO:Ni than in pure MgO, besides the relative intensity of the 357 and 570 nm bands is different. The total intensity of the absorption bands from the sample irradiated in electric field is lower than that from those irradiated without electric field. The electric field changes the rate of annealing. Differences between the annealing curves for pure and nickel-doped MgO indicate that the nature of defects is different. We assume that the 347 nm and 480 nm absorption bands detected after annealing belong to complex Ni^{3+}-V_{Mg}.

1. INTRODUCTION

Investigating radiation resistance of materials for future fusion application, it is very important to understand the mechanisms of damage formation under irradiation. During the last few years a considerable work has been carried out on study of thermally stimulated impurity diffusion and aggregation in MgO[1-4]. However there is little information on the influence of radiation on impurity diffusion[5-6], especially under influence of electrical field[7]. Optical absorption measurement yield useful information on the charge trapping and recombination center structure in irradiated samples.

The paper presents a study of effect of electric field on transport of radiation defects in pure and nickel- doped MgO.

2. EXPERIMENTAL DETAILS

MgO crystals used in this investigation have been grown by the arc fusion method in Russia, the single solid solutions MgO-NiO are grown by the chemical transport reaction method[8]. MgO crystals are used as substrates for single solid solution growth. The sample's thickness was in the range of 0.5–1 mm.

The impurities concentration was detected by means of the instrumental neutron activation analysis[9]. The results are presented in the table 1.

The neutron irradiation was fulfilled at Latvian 5MW water- water research reactor. The fluence of fast neutrons with energy > 0.1 MeV was in the range $10^{14} - 5 \cdot 10^{18}$ cm^{-2}, that of thermal neutrons - 10^{14} - 2.5 10^{17} cm^{-2}. The accompanied γ-irradiation dose with average energy 1.1 MeV gave an absorption dose of 0.33 Gy. The range of irradiation temperature was 300 – 700 K. Cadmium filter was used for absorption of thermal neutrons. The samples were placed between two plates of nickel electrodes like sandwich and located in the resistance furnace that allowed to change temperature during irradiation up to 700 K. The electric field was 10 kV/m large. Direction of the electric field was perpendicular to neutron flux and coincided with to the crystal plane (100). The thermal evolution of the absorption bands was observed at isochronal annealing during 10 minutes at each temperature.

Table 1. Concentration of impurities (in mass%)

Notation	Cr	Fe	Mn	Ni
MgO 1	1.75×10^{-4}	1.1×10^{-2}	5.8×10^{-3}	$<5 \times 10^{-5}$
MgO 2	8.5×10^{-4}	7.3×10^{-4}	$< 10^{-5}$	-
MgO 3	1.8×10^{-4}	4.4×10^{-2}	2.4×10^{-4}	$< 5 \times 10^{-5}$
MgO 4	2.11×10^{-4}	8.1×10^{-3}	1×10^{-3}	-
MgO 5	3.7×10^{-3}	7.3×10^{-3}	5.6×10^{-3}	-
MgO:Ni	2×10^{-4}	1×10^{-2}	-	0.35
MgO:Ni	2×10^{-4}	7×10^{-3}	$< 10^{-5}$	0.51
MgO Ni	6.9×10^{-5}	5.1×10^{-4}	2.7×10^{-4}	1.7×10^{-4}

The table 2 presents the results of the determination of the lattice parameters in the pure and nickel- doped magnesium oxide. The X-ray diffraction spectra were measured by diffractometer DRON-UM2 (USSR) using CuKα-radiation (U=40 kV, I=20 mA). A computer connected with the diffractometer was used for logging and processing data.

Table 2. Lattice parameters before and after neutron irradiation (a, nm)

Notation	MgO	MgO:Ni	MgO:Ni	MgO:Ni	NiO
before irradiation	0.4211	0.4208	0.4209	0.42102	0.4170
after fast $\Phi=10^{17}cm^{-2}$	-	0.4250	0.42055	0.42088	-
thermal $\Phi=2.5 \cdot 10^{17}$ cm^{-2}	-	-	-	0.42043	-
after annealing	-	0.4286	-	-	-

The conventional technique used to measure absorption spectra was based on "Specord M-40" (Karl Zeiss Jena) double-beam spectrophotometer operating in the wavelength region of 50000–11000 cm^{-1} (200–900 nm). Optical measurements before and after irradiation were made at room temperature.

3. RESULTS AND DISCUSSION

It is known that absorption spectra of neutron irradiated pure MgO consists of three bands associated with radiation defects[10-13]. In additional to radiation defects the small content of nickel impurity in octahedral sites rises absorption bands due to the spin-allowed transition[14] from the ground state $^3A_{2g}(^3F)$ to the exited states $^3T_{2g}(^3F)$, $^3T_{1g}(^3F)$ and $^3T_{1g}(^3P)$ (Fig. 1, curve 1). At sufficiently large impurity concentration (c ~ 0.1 mass.%) the accumulation of radiation defects differs from that in pure MgO crystals.

The intensity of the ~570 nm absorption band is considerably smaller in MgO: Ni than in pure MgO crystals under the same fluence of the fast neutrons (Fig. 1, curve 5 and Fig. 1, curve 1). Besides the intensity ratio of the 357 and 570 nm bands is also different. For pure MgO single crystals the intensity of the ~570 nm band is 20 times larger than intensity of the 357 nm band. In the case of nickel-doped MgO the intensities of these bands are approximately equal.

The behaviour of absorption bands during isochronal annealing in pure and doped crystals is also different. For MgO: Ni intensity of the ~570 nm band doesn't change up to 700 K and then sharply decreases and disappears at 900 K (Fig.2). While in pure MgO intensity of the ~570 nm band slowly decreases (Fig. 3) up to 650 K then increases up to 900 K. However the annealing curve for MgO substrate separated from

$Ni_{0.125}Mg_{0.875}O$ single solid solution after irradiation has the same behaviour as for MgO: Ni (Fig.3, curve 3).

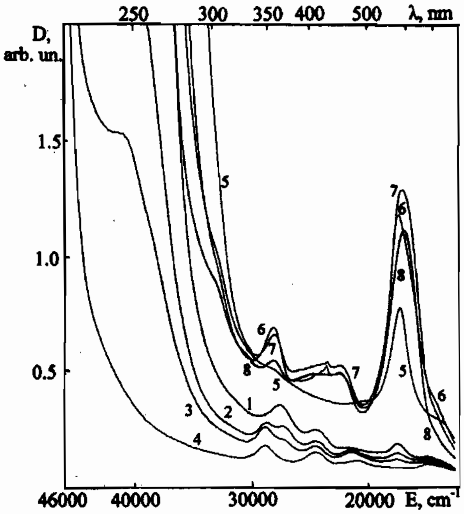

FIGURE 1
Absorption spectra of crystals irradiated of fast neutron fluence $10^{18}cm^{-2}$: 1. MgO:Ni; 5. MgO; 2. MgO:Ni annealed at 660 K; 3. at 730 K; 4. at 878 K; 6. MgO annealed at 790 K; 7. at 850 K; 8. at 915 K.

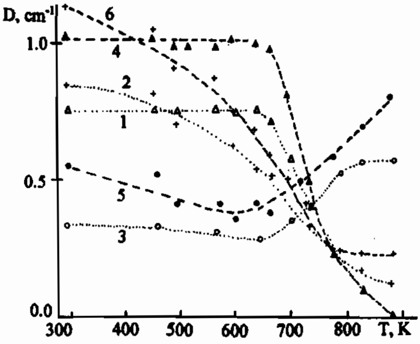

FIGURE 2
Absorption band intensities of MgO:Ni as functions of annealing temperature: 1. 570 nm; 2. 357 nm; 3. 347 nm in electric fields; 4. 570 nm; 5. 347 nm; 6. 357 nm without electric field

Annealing of the 357 nm band depends on a sample type. For pure MgO intensity of the band doesn't change up to 500 K then increases, reaching maximum at 750 K and disappears at T>900 K (Fig. 4). The annealing of MgO: Ni crystals shows that the 357 nm band consists of two band: 362 and 347 nm (Fig. 1, curve 2). The intensity of the 362 nm band decreases and that of the 347 nm band increases during annealing. In the same time we observed the appearance of the 480 nm band and disappearance of the 460 nm band.

Fig. 5 and Fig. 6 shows the absorption spectra of pure and nickel-doped MgO under and without electric field. The total intensity of the absorption bands from the sample irradiated in electric field is lower than that from those irradiated without electric field. Annealing in electric field of the samples irradiated without electric field shows difference in intensity of the ~570 nm band at temperature higher than 800 K for pure and nickel-doped MgO (Fig. 2 and. Fig. 3.).

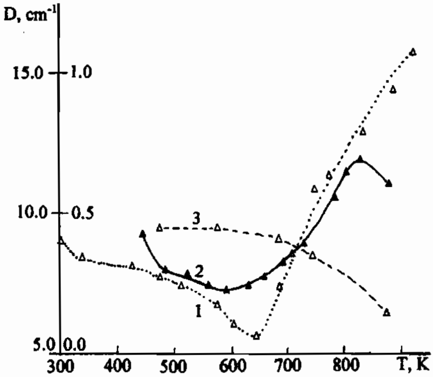

FIGURE 3
Absorption band 570 nm intensities of MgO as functions of annealing temperature: 1. without electric field; 2. in electric fields; 3. MgO substrate without electric field.

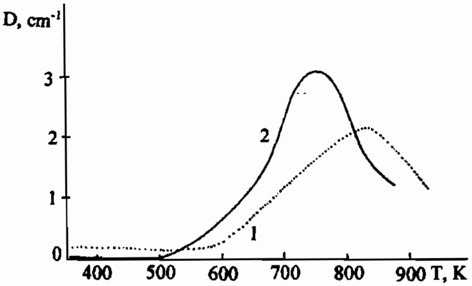

FIGURE 4
Absorption band 357 nm intensities of MgO as functions of annealing temperature: 1. without electric field; 2. in electric fields.

As indicated previously[15] impurities and their concentration play an important role in determining the lifetime of MgO crystals against electrothermal breakdown. The presence of Fe, Ni, Cr, or V enhances the breakdown. The increase of current results from the increase of the hole injection, which is enhanced by accumulation of negative charged ionic species or vacancies near the anode, as well as development of the local electric neutrality. In the case of vacancy migration, the migration of defects (and hence the charge) proceeds via a random series of jumps[16]. The electric field changes the rate of annealing.

Differences between the annealing curves for pure and nickel-doped MgO indicate that the nature of defects is different. During irradiation of crystals, the radiation energy is mainly dissipated on excitation of the electron subsystem of the crystal, i.e. on ionizing. Ionization processes may lead to a change of a charge of genetic defects and formation of metastable defect centers including color centers. A part of radiation energy is lost in elastic collisions of radiation particles or secondary ones with crystal atoms. In irradiation process as a result of elastic collision the nickel ions can form the complex centers, which consist of vacancies and nickel ions. The 570 nm band is assigned to the aggregated F-centres. This explains why the intensity of the 570 nm band in MgO:Ni is considerably smaller than that in pure MgO, i.e. a part of anion vacancy (F^+-center) capture nickel ions and form complexes "Ni^{2+}-F^+-center".

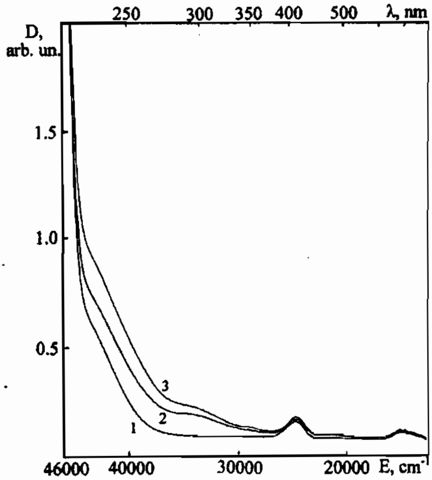

FIGURE 5
Absorption spectra of MgO:Ni 1. before irradiation; 2. after fast neutron irradiation at fluence $10^{17}cm^{-2}$ in electric field; 3. after irradiation at fluence $10^{17}cm^{-2}$ without field.

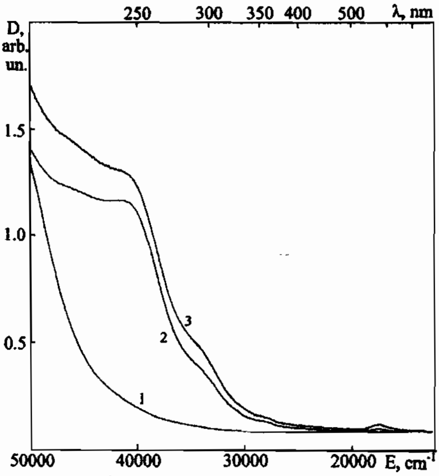

FIGURE 6
Absorption spectra of MgO 1. before irradiation; 2. after fast neutron irradiation at fluence $10^{17}cm^{-2}$ in electric field; 3. after irradiation at fluence $10^{17}cm^{-2}$ without field.

It is known[17], that in closed-shell oxides like MgO a hole may change the charge state of one of the neighbouring transition metal cations. In the ground state, the hole may localize and self-trap on one of the equivalent ions like six O neighbours of a Mg vacancy in MgO, forming an O^- center. For MgO there are intra-ionic excitations and charge transfer transitions in a result of which the hole is transferred from the O^- to another oxygen, or to some group of equivalent oxygen's. The intra-ionic transition energy is largely determined by the crystal field, the electric field at the O^- dominated by the cation vacancy. A change of the valence state from Ni^{2+} to Ni^{3+} is possible according to the equation: $Ni^{2+} + O^{-} \rightarrow Ni^{3+} + O^{2-}$. The charge is compensated by positive ion vacancies. We assume that the 347 nm and 480 nm absorption bands detected after annealing belong to such complex Ni^{3+}-V_{Mg}. The experiments on electron paramagnetic resonance show the signal attributed to Ni^{3+}ions for such annealed crystals[18]. The changes of lattice parameters observed in the irradiated NiO[19] are explained by the creation of Ni^{3+}. After neutron irradiation we also observe a change in the lattice parameters (Tab. 2) confirming our suggestion.

4. CONCLUSION

It is found that the accumulation of radiation defects in nickel-doped MgO differs from that in pure MgO. The intensity of the 570 nm absorption band is considerable smaller in MgO:Ni than in pure MgO, besides the relative intensity of the 357 and 570 nm bands is different. The change in intensity of the absorption bands is more pronounced in MgO with nickel impurities. Differences between the annealing curves for pure and nickel-doped MgO indicate that the nature of defects is different. The total intensity of the absorption bands from the sample irradiated in electric field is lower than that from those irradiated without electric field. The electric field changes the rate of annealing. We assume that the 347 nm and 480 nm absorption bands detected after annealing belong to complex Ni^{3+}-V_{Mg}.

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