INTERACTION OF NUCLEAR RADIATIONS WITH SCINTILLATION CRYSTALS ZnSe(O, Te)

L.P. Gal'chinetskii, V.D. Ryzhikov, N.G. Starzhinskiy
Institute for Scintillation Materials of NAS of Ukraine
60 Lenin Ave, Kharkov 61001, Ukraine
E-mail: stcri@isc.kharkov.com

The experimental data on the irradiation of ZnSe(O,Te)-crystals with γ-quanta, protons and neutrons are presented and discussed. The mechanism for heightening the concentration of luminescence centers of C1-type $Zn_iV_{Zn}Te_{Se}$ ($\lambda_{max}\approx640$ nm) and C2-type $V_{Zn}Zn_iO_{Se}$ ($\lambda_{max}\approx605$ nm), accompanied by an increase in the radioluminescence intensity, is submitted. During p-irradiation, hydrogen, generated in the process of proton thermalization, interacts with oxygen that the C2-centers contain, which results in the C2-center destruction, a decrease in the intensity and the λ_{max} shift towards the long-wave range.

PACS: 29. 27. Fa

1. INTRODUCTION

The scintillation crystal ZnSe(O,Te) is characterized by its combination of characteristics making it the optimum choice for "scintillator-photodiode" detectors. The most important parameters of ZnSe(O,Te) crystals are presented in Table in comparison with CsI(Tl).

Principal parameters of scintillators ZnSe(O,Te)

Parameter	Scintillator material		
	CsI(Tl)	ZnSe(O,Te)	
		"fast"	"slow"
Melting temperature, K	894	1773…1793	1773…1793
Density ρ, g/cm^3	4.51	5.42	5.42
Effective atomic number, Z	54	33	33
Hygroscopicity	low	no	no
Emission maximum, λ_{max}, nm	550	605	640
Afterglow, δ (after 6 ms), %	0.1…5.0	<0.05	<0.05
Attenuation coefficient of intrinsic radiation (λ_{max}=605…640 nm), α, cm^{-1}	<0.05	0.05…0.2	0.05…0.2
Light yield from PD with respect to CsI(TI) at 1 mm thickness, E_x=60 keV, %	100	up to ~110	up to ~140
Decay time, τ, μs	1	1…3	30…70
Maximum value of the spectral matching factor K_u	0.77	0.9	0.92
Refraction coefficient, n	1.79	2.61	2.59
Light yield, photons/MeV-γ	5.5…10^4	up to 6· 10^4	up to 7.5· 10^4
Depth of 90% absorption X-ray (40 keV), mm	<0.25	0.65	0.65

It is clearly seen that ZnSe(O,Te) is obviously superior in such qualities of primary importance as conversion efficiency and absolute light output, afterglow, spectral matching, hygroscopicity. In addition, absorption depth in CsI(Tl) at energies of less than 40 keV is less than 0.25 mm. This means that negative influence of the surface layer upon scintillation parameters is negligible with ZnSe(O,Te), but it is significant with thinner CsI(Tl) samples. Thus, advantages of ZnSe(O,Te) in comparison with CsI(Tl) as scintillator for low-energy detector arrays are clear and undeniable. Among advantages of ZnSe(O,Te), one should also note its high radiation stability (not less than 10^7 rad, γ-,X-irradiation). All this shows that ZnSe(O,Te) is the most promising material for "scintillator-photodiode" detectors of γ-, X-radiation in the range of up to 40…60 keV.

In the given paper the influence of ZnSe(O,Te) crystal irradiation with neutrons and protons on the luminescent and optical characteristics of these crystals is examined.

2. EXPERIMENTAL RESULTS AND DISCUSSION

The experimental conditions have been the following. Gamma-irradiation: a channel type ^{60}Co, exposure dose rate P_γ up to $3\cdot10^3$ R·s^{-1} (the average energy of gamma-quanta $E_\gamma\approx1.25$ MeV), the absorbed dose $D\gamma\leq8.5\cdot10^8$ rad. Proton irradiation (p): U-150 type cyclotron (E_p=18 MeV; beam current density $1.9\cdot10^8$ A·cm^{-2}; fluence $F_p\leq1.7\cdot10^{15}$ cm^{-2}). Neutron (n) irradiation: the thermal channel of a WWR-SM type nuclear reactor at $P_{tn}=1.2\cdot10^{11}$ neutron·cm^{-2}·s^{-1}, the fluence values of $F_{tn}=1.3\cdot10^{16}$ neutron·cm^{-2} (cadmium ratio >6). X-ray luminescence (XL) of semiconductor scintillator samples was measured under excitation using an IRIS-3 X-ray source (U_a=35 kV, $i_a\leq35$ mA, Cu–anticathode).

Under irradiation of all these types, the temperature of the samples did not exceed 360 K.

In Fig.1 XL-spectra of ZnSe(O,Te) samples after neutron (n-) and proton (p-) irradiation are presented. As Fig.1 indicates, the spectrum remains unchanged after n- irradiation, whereas p- irradiation causes the spectrum shift towards the long- wave region.

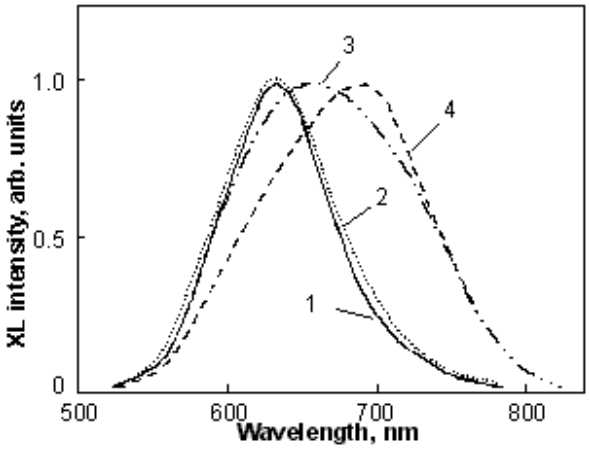

Fig.1. XL spectra of ZnSe(O,Te) samples: 1 – unirradiated; 2 – after neutron irradiation (F_{nt}=1.3·10^{16} neutrons·cm^{-2}); 3, 4 – after proton irradiation (F_p=1.2·10^{14} protons·cm^{-2} and 1.7·10^{15} protons·cm^{-2}, respectively)

In Fig.2 the transmission spectra of these samples in IR region are depicted.

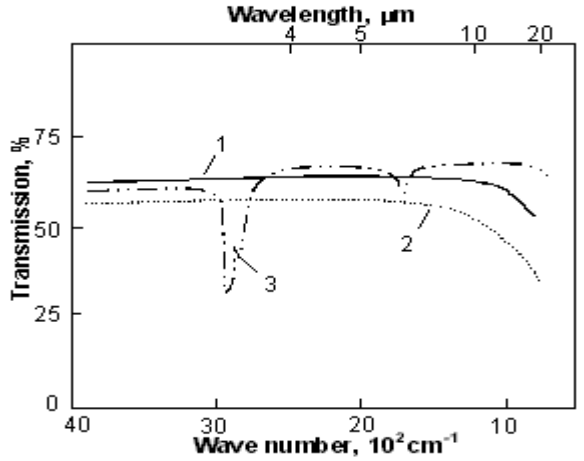

Fig.2. IR absorption spectra: 1 – unirradiated ZnSe(O,Te) samples; 2 – after neutron irradiation (F_{nt}=1.2·10^{14} neutrons·cm^{-2}); 3 – after proton irradiation (F_p=1.7·10^{15} protons·cm^{-2})

After n- irradiation the nonselective absorption remains almost unchanged in the visible range (λ >600 nm), and it essentially increases in the IR range (see Fig. 2).

After p- irradiation in the visible range (λ>600 nm) the nonselective absorption increases approximately by 20%, and it diminishes in the IR range (see Fig.2). However, there arise selective absorption peaks when the wave number makes ≈3000 cm^{-1} and ≈ 1600 cm^{-1}. Besides, after n- irradiation (F_n= 1.3·10^{16} neutrons·cm^{-2}) the X-ray luminescence intensity (I_{XL}) increases by 20…50%, while the γ- luminescence intensity (I_{GL}) increases by 10…300%, and dark electrical conductivity increases by order of magnitude.

After p- irradiation (F_p>10^{15} protons·cm^{-2}) the X-ray luminescence intensity (I_{XL}) decreases almost by 90%, I_{GL} becomes lower almost by 95%, electric conductivity diminishing by 1.5…2 orders of magnitude.

In Fig.3 the temperature dependences of ZnSe(O,Te) crystal I_{GL} are depicted under irradiation with the same doses of neutrons and protons.

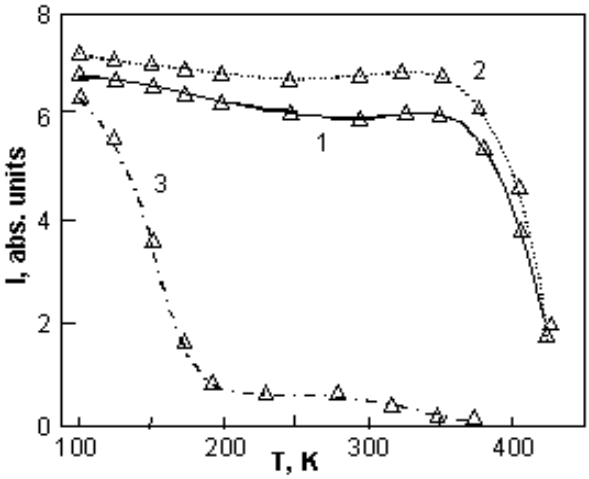

Fig.3. Temperature dependence of I_γ for ZnSe(O,Te) crystals irradiated by neutrons (2) and protons (3); 1 – initial sample

As Fig.3 indicates, n- irradiation does not cause any essential change in I_{GL} as compared with the unirradiated sample. At the same time, p- irradiation causes the substantial changes in the temperature dependence of I_{GL} as compared with the unirradiated sample.

In order to explain these data, the use should be made of the investigations of the mechanism for the radiative recombination (RR) in ZnSe(O,Te) crystals within the range 600…640 nm. Earlier the structure of radiative recombination centers (RRC) in these crystals has been studied (see [1,2]). As it is found out, in ZnSe crystals doped with isoelectronic admixtures – oxygen (O_{Se}) or tellurium (Te_{Se}) – RRC have the form of the complexes V_{Zn} Zn_i O_{Se} (luminescence at λ_{max} ≈ 605 nm) or Zn_i V_{Zn} Te_{Se} (λ_{max} ≈ 640 nm). In [1,2] it is also demonstrated that the real content of these RRC is close to the free carrier concentration (N_e). The latter can be estimated by Drude formula on the basis of measurements of the absorption in that region of IR range where it is conditioned by free carriers [3]. According to these estimations, in ZnSe(O_{Se}, Te_{Se}) crystals N_e<2·10^{16} cm^{-3}. Under the real conditions the crystals are doped with tellurium in the amount ~ 10^{20} cm^{-3}, introduced in the form of the solid solution (SS) of $ZnSe_{1-x}Te_x$. Thus, just <0.1% of the tellurium amount that gets into the crystal has the form of Te_{Se}. The major amount of Te makes a component of the metastable SS and it also can be accumulated at dislocations.

This reasoning allows us to suppose that in the case of n- irradiation the effects of increase in I_{XL}, I_{GL} and increase in the absorption within IR range are induced by the radiation- stimulated processes of the Te_{Se}, O_{Se} concentration increase in ZnSe(O,Te) crystals. These processes are accompanied by increase in the amount of RRC-1 (λ_{max} ≈ 640 nm) and RRC-2 (λ_{max} ≈ 605 nm).

Let's dwell on explaining the results of the experiments on p- irradiation. It is proved that ZnSe(O,Te) crystal thermal treatment in the hydrogen atmosphere causes either destruction or transformation of RRC-2 according to the following schemes:

$$H_2 + V_{Zn}\, Zn_i\, O_{Se} \rightarrow Zn_{Zn} + H_2O\uparrow \quad (1)$$

$$H_2 + V_{Zn}\, Zn_i\, O_{Se} \rightarrow V_{Zn}\, Zn_i + H_2O\uparrow \quad (2)$$

These processes both cause a decrease in the radioluminescence intensity and the luminescence spectrum λ_{max} shift towards the long- wave region.

Consequently, decrease in I_{XL}, I_{GL} and the absorption diminution within IR range as well as the XL λ_{max} shift, detected after p- irradiation, are explicable in the following way. In the course of ZnSe(O,Te) crystal p- irradiation hydrogen, generated during the proton thermalization, interacts with oxygen, present in RRC-2. This results in destruction or transformation of RRC-2 according to the schemes (1) and (2), respectively. The existence of this hypothetical mechanism is confirmed by the fact that the selective absorption in vicinity to 3000 cm^{-1} (see Fig.2) can be conditioned by valence oscillations of the groups O – H or O – D, whereas the absorption in vicinity to 1600 cm^{-1} can be caused by deformation oscillations of H_2O molecule.

3. CONCLUSION

Increase in the radioluminescence intensity of ZnSe(O, Te) crystals during n- irradiation is conditioned by radiation- stimulated processes in the crystals, accompanied by augmentation in concentrations of RRC-1 and RRC-2.

In the course of p- irradiation, hydrogen, generated during the proton thermalization, interacts with oxygen, present in RRC-2, which results in the RRC-2 destruction, diminution in RL intensity and λ_{max} shift towards the long- wave region.

4. ACKNOWLEDGMENT

The authors are grateful to Magne Guttormsen (Oslo, Cyclotron Laboratory, Norway) and Alexander A. Kist (Institute Nuclear of Physics, Uzbekistan) for providing the sample irradiation. Besides, the authors express their gratitude to V.A. Tarasov and A.I. Mitichkin (ISMA, Kharkov, Ukraine) for the useful discussion of the results.

REFERENCES

1. L.V. Atroschenko, S.F. Burachas, L.P.Gal'chinetskii et al. *Crystals of scintillators and detectors of ionizing radiations on their base*. Kiev: "Naukova dumka", 1998, p.312.
2. V.M. Koshkin, A.Ya. Dulfan, V.D. Ryzhikov et al. Thermodynamics of isovalent tellurium substitution for selenium in ZnSe semiconductors // *Functional materials*. 2001, v.8, №4, p.708-713.
3. V. Ryzhikov, N. Starzhinskii, K. Katrunov et al. Effects of annealing in zinc vapor upon the electron spectrum formation of ZnSe(Te) – based scintillators // *Functional materials*. 2002, v.9, №1, p.143-146.

ВЗАИМОДЕЙСТВИЕ ЯДЕРНЫХ ИЗЛУЧЕНИЙ СО СЦИНТИЛЛЯЦИОННЫМИ КРИСТАЛЛАМИ ZnSe(O,Te)

Л.П. Гальчинецкий, В.Д. Рыжиков, Н.Г. Старжинский

Приведены результаты экспериментов по облучению кристаллов ZnSe(O,Te) γ-квантами, протонами, нейтронами. Предложен механизм увеличения при n-облучении концентрации центров люминесценции Ц1 типа $Zn_iV_{Zn}Te_{Se}$ (λ_{max}≈640 нм) и Ц2 типа $V_{Zn}Zn_iO_{Se}$ (λ_{max}≈605 нм), сопровождающий возрастанием интенсивности I радиолюминесценции (РЛ). При p-облучении водород, образующийся в процессе термализации протонов, взаимодействует с кислородом, входящим в состав Ц2; это приводит к разрушению Ц2, уменьшению I РЛ и к длинноволновому сдвигу λ_{max}.

ВЗАЄМОДІЯ ЯДЕРНИХ ВИПРОМІНЮВАНЬ ЗІ СЦИНТІЛЯЦІЙНИМИ КРИСТАЛАМИ ZnSe(O,Te)

Л.П. Гальчінецький, В.Д. Рижиков, Н.Г. Старжинський

Наведено результати експериментів з опромінення кристалів ZnSe(O,Te) γ-квантами, протонами, нейтронами. Запропоновано механізм збільшення під час n-опромінення концентрації центрів люмінесценції Ц1 типу $Zn_iV_{Zn}Te_{Se}$ (λ_{max}≈640 нм) та Ц2 типу $V_{Zn}Zn_iO_{Se}$ (λ_{max}≈605 нм), якиї супроводжується зростанням інтенсивності I радіолюмінесценції (РЛ). Під час p-опромінення водень, що утворюється в процесі термалізації протонів, взаємодіє з киснем, який входить до складу Ц2; це призводить до зруйнування Ц2, зменшення I РЛ та до довгохвильового зсуву λ_{max}.