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Investigation of luminescent properties of ^{6}Li-based fibers at soft X-ray excitation

I N Sedunova[1], V Yu Ivanov[1], V I Arbuzov[2], K V Dukelsky[2], B V Shulgin[1], S I Gorkunova[1], V N Churmanov[1] and I L Vitovsky[1]

[1] Yeltsin Ural Federal University, Mira St. 19, 620002, Yekaterinburg, Russia
[2] Vavilov State Optical Institute, Srtiom, Babushkina Str. 36/1, 193171, Saint-Petersburg, Russia

E-mail: i.n.sedunova@gmail.com

Abstract. The investigation of luminescent and scintillation properties of $^6Li_20$-MgO-SiO_2-Ce fibers is presented. The probable mechanisms of energy transfer to activator centers are discussed. The estimation of fibers absolute scintillation efficiency was implemented. The modeling of neutron registration efficiency in substitution of monocrystalline detector by a fiber one was carried out.

1. Introduction

The neutron detection presents an urgent problem for different applications: radiation monitoring, neutron spectroscopy, industrial and medical measurements, health physics, etc. Most neutron detection devices presently are based on ^{3}He gas-filled detectors. They are used especially for low neutron fluxes detection and have excellent neutron-to-gamma discrimination. At the same time the scintillation method is widespread for large neutron fluxes detection.

The scintillation material $^6Li_2O$-MgO-SiO_2-Ce^{3+} intended for registration of thermal neutrons was developed at the Vavilov State Optical Institute. This compound has a high radiation resistance, light yield and neutron sensitivity. Thermal neutrons are detected by the $^6Li(n,\alpha)^3H$ reaction. The reaction products – α-particles – cause 40-60 ns scintillation in glass. The investigation of luminescent and scintillation characteristics of this material in a bulk form was carried out early [1,2].

The present work is devoted to studying the properties of ^{6}Li-based fibers produced at the Vavilov State Optical Institute. Applied investigation of the similar fibers was reviewed in [3]. The substitution of monocrystalline detector by a fiber one must be advantageous because the fibers using leads to decreasing the light loss. In addition, the fiber technology has a lot of advantages: wide range of detector sizes (from single fiber to large area detector), flexibility and conformability allowing optimization of the source to detector geometry.

2. ^{6}Li-based fibers producing

The ^{6}Li-fibers were produced using following technology. The melting of scintillation silicate glass doped with cerium and contained 22.5 mol.% of Li_2O was carried out. High purity reagents including Li_2CO_3 enriched in ^{6}Li (85 %) were exploited. Glass-ceramic crucible charged with mixture of oxides and salts was placed in the laboratory furnace with silicon carbide heaters. The glass melting was implemented in reducing atmosphere at 1450ºC for 6-8 hours with glass bath mixing by means of thimble of the same material as crucible one. Following melting the glass bath was filled in graphite mold which then was placed in muffle furnace where the temperature decreased up to room one. Annealed glass plates with 10 mm thickness and 80 mm width was polished and then cut up to special material blocks with 30-40 cm length and 5x5 mm^2 section for fibers producing. Side surfaces of these blocks were polished to exclude the crystallization of fibers surface layers.

The heater temperature was selected so that glass viscosity in softening zone was not more than 10^7 Pa·s, but below temperature of block material crystallization. After the optimal temperature conditions were chosen the fibers producing was carried out. Thereto the blocks were put in resistance furnace with graphite heater in the form of a cylindrical ring. Purified argon flow was run through the internal space of heater.

Fibers were produced at a rate of 0.5 m/min. About 10 meters of fiber element with 400x400µm^2 section was synthesized from one material block.

3. Experimental technique and methods

Luminescence spectra (LS) (2.5 – 5 eV), luminescence excitation spectra (LES) (45-250 eV) and luminescence kinetics at T=10 K and 300 K were measured on BW3 channel of DORIS synchrotron (HASYLAB, DESY, Hamburg). The luminescence was excited using Zeiss SX700 monochromator (average spectral resolution is 0.04 eV for energy range of 45-250 eV). The LES were corrected for the equal number of the exciting photons. LS at 2.5-5 eV was registered by means of 0.4 m vacuum Seya-Namioka monochromator coupled to MCP 1645 (Hamamatsu).

Optical absorption spectra of ^{6}Li-based fibers were measured at T=300 K using UV-Visible Helios Alpha spectrophotometer. The pulse excitation with electrons of 140 keV was carried out using MIRA-2D pulsed source.

Scintillation efficiency was assessed using fibers irradiation with α-particles from ^{239}Pu-sourse (activity – 40 kBq, average particle energies – 5.1 MeV). Electronic signal from PMT-130 working in current mode passed to spectrometric amplifier and then to ADC board of built-in multichannel spectrometer manufactured by LTD «ASPECT» (Dubna, Russia).

4. Experimental results

Luminescence spectrum of ^{6}Li-based fiber at the excitation by 130 eV photons is presented in figure 1 and is not elementary one. Satisfactory decomposition on Gaussian components was carried out only by four fitting curves (inset in figure 1). Taking into account that luminescence derived from d-f transitions in cerium, we should conclude the existence of at least two non-equivalent Ce^{3+} positions. The analysis of luminescence excitation spectra profile (figure 2) allows to discuss the mechanisms of energy transfer in ^{6}Li-based fibers. We have observed a well-outlined structure at the energy range of 100-135 eV, and the first resonance has the energy more than the one of L-edge of absorption of silicon ions (99.42 eV for free ion). The observed structure was investigated for silicates and was associated with the presence of unfilled molecular orbitals of different type (s-, p- and d-type) in silicon-oxygen tetrahedron [4]. So well-outlined resonance at L-edge of absorption of silicon ions was firstly observed for Si-contained materials and it testify to a high efficiency of energy transfer channel «silicon-oxygen tetrahedrons→Ce».

The decay kinetic (figure 3) measured for 3.2 eV at 130 eV excitation includes several components – the extremely short (<1 ns and 4.5 ns) and the one of 26 ns. The latter decay time is likely to be associated with Ce^{3+} emission. The presence of short decay times may indicate the manifestation of different cerium cites. The investigation of non-equivalent positions of cerium was reviewed in [5]. The presence of short decay times gives rise to consider ^{6}Li-based fibers as effective scintillator in terms of temporal resolution. But under pulse excitation with electrons of 140 keV the decay times increased up to 83 ns.

The results of optical absorption and luminescence spectra measurements are shown in figure 4. The measurement results indicate that the long-wavelength edge of absorption spectra and cerium emission spectra are overlapped. Therefore, partial reabsorption of impurity luminescence is probable,

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and we should consider that as disadvantage in terms of using this material as efficient scintillator. To shift the optical absorption spectrum to short-wavelength region we proposed to vary the chemical mixture of ^{6}Li-based fibers.

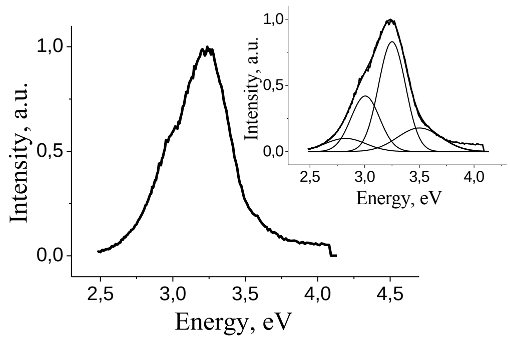

Figure 1. Luminescence spectra of ^{6}Li-based fibers at E_{ex}=130 eV, T=300 K.

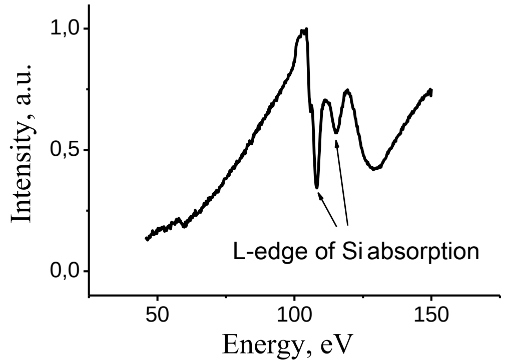

Figure 2. Luminescence excitation spectra of ^{6}Li-based fibers for E_{em}=3.2 eV, T = 300 K.

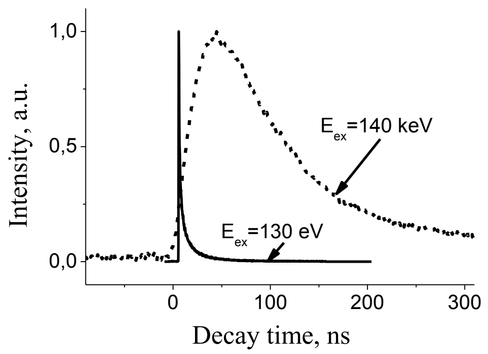

Figure 3. Luminescence kinetics of ^{6}Li-based fibers for E_{em}=3.2 eV at E_{ex}=138 eV and E_{ex}=140 keV, T=300 K.

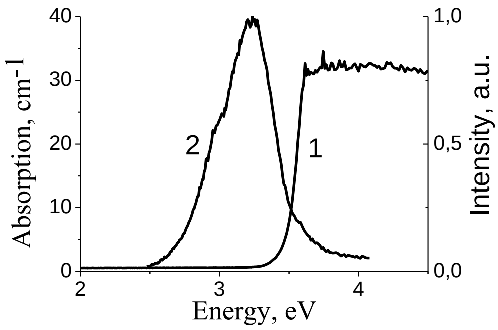

Figure 4. Comparison of absorption (1) and luminescence (2) spectra of ^{6}Li-based fibers at T=300 K.

5. Modelling and calculations

For materials considered as efficient scintillators it is important to estimate absolute scintillation efficiency. The measured absolute scintillation efficiency for ^{6}Li-based fibers was 1.10%.

To find optimal construction parameters of scintillation detector we have carried out the modeling of neutron registration efficiency and the estimation of light collection coefficient using Monte-Carlo method in Delphi 7. The registration efficiency is considered as a ratio of neutron registered to ones fell on scintillator. The light collection coefficient is as ratio of photons fallen on photoelectric cathode to photons emitted in scintillator. As we suppose the substitution of monocrystalline detector by fiber one must be advantageous because the fibers using leads to light transfer by means of internal reflectance and the decreasing the light loss, consequently.

We have designed a model taking into account the following points: isotropic material, straight-line pass of light, consideration of neutron-capture reaction on ^{6}Li nuclei. The schematic view of reference construction describing neutron registration by using monocrystalline detector is presented in figure 5, the ultimate construction with fiber detector (and without light guide) – in figure 6.

Program algorithm includes the consideration of the following events:

- Neutron generation with a glance on its energy and direction vector.
- Calculation of collision with moderating material points (x0,y0,z0) and determination of the neutron energy and direction vector on each step until the particle gets to detector.
- Consideration of the interaction of particle and detector leading to scintillation. The photon number depends on absolute scintillation efficiency of material.
- Consideration of the photon propagation up to photoelectronic multiplier.

On the base of this algorithm the neutron registration efficiency and light collection coefficient are calculated. The modeling results are presented in table 1.

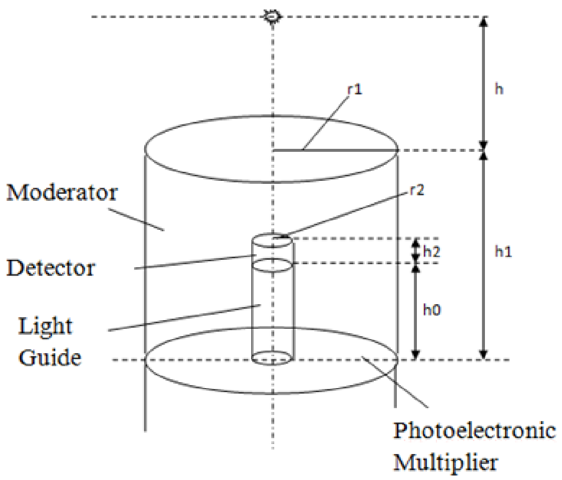

Figure 5. Monocristalline detector.

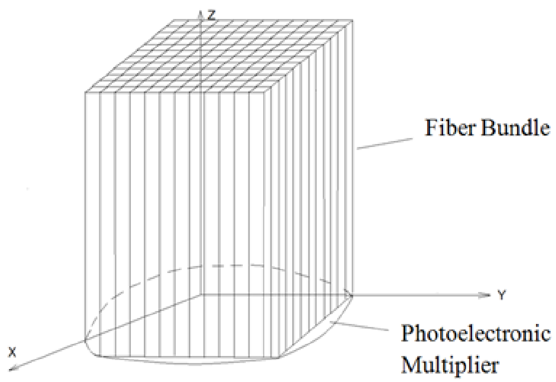

Figure 6. Fiber detector.

Table 1. Results of modelling.

Fiber bundle length (cm)	Light guide length (cm)	Light collection coefficient of bulk glass (%)	Light collection coefficient of fiber (%)	Neutron registration efficiency (%)
0.6	4.2	0.57	2.48	4.16
1.2	3.6	0.57	7.36	4.04
2.4	2.4	1.10	9.94	4.01
3.6	1.2	2.70	10.1	3.83
4.8	0.0	13.9	18.2	3.71

The modeling has shown the fiber bundle result in increasing of light collection coefficient by several times in comparison with monocrystalline detector. We interpret that phenomenon as the decreasing of light loss in photon transport to photoelectronic multiplier due to internal reflectance observed in fibers. The low values of light collection coefficient for bulk glass can be explained by the fact of partial reabsorption in glass (figure 4). The relatively high values in case of light guide absence are connected with the decreasing of optical contacts in construction and the decreasing of light losses due to absorption in light guide (the absorption coefficient is about 90 m^{-1})

6. Conclusion

The comprehensive investigation of luminescent and scintillation properties of $^6Li_2O$-MgO-SiO_2-Ce^{3+} fibers was carried out. The possible mechanisms of energy transfer to cerium centers are discussed. We suppose that probable mechanism is the «silicon-oxygen tetrahedrons→Ce» energy transfer channel. The estimation of absolute scintillation efficiency was carried out. The modeling of monocrystalline detector substitution by fiber has shown that the latter configuration is more advantageous.

Acknowledgements

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