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PERSPECTIVES OF PURE AND Ce^{3+}-DOPED SrF_2 CRYSTALS IN SCINTILLATION APPLICATIONS[1]

In this paper results of measurements of scintillation properties of pure and Ce^{3+}-doped crystals are presented. Light yield of these materials is compared with the light output of well-known scintillators – NaI–Tl and CaF_2–Eu. The crystals of SrF_2–Ce^{3+} have high light yield and they are perspective scintillators for well-logging detectors.

Keywords: *scintillators, light output, fluroide, cerium doped.*

1. Introduction

The interest in new scintillation materials is promoted by increasing number of new applications in medicine, science, and homeland security, which require ramp-up of material production. The most perspective scintillators are bromides and iodides doped with Ce^{3+} and Eu^{2+} ions, such as SrI_2–Eu and $LaBr_3$–Ce. These crystals have high light yields (up to 100000 ph/MeV for SrI_2–Eu), good energy resolution, and high proportionality [1]. Disadvantages of these scintillators are high hygroscopicity and price. In addition, SrI_2–Eu has long decay time (about 2 μs) and temperature instability of light yield [2].

For the most applications a cheaper NaI–Tl scintillator has quite properties (light yield about 45000 ph/MeV [3]). Therefore, one of the way in development of new scintillators is to find new materials with close to NaI–Tl properties but no hygroscopic. In this way a perspective materials for new scintillators are alkali-earth fluorides doped with rare earth ions. Theoretical limit of light yield calculated for these materials is up to 50000 ph/MeV [1]. If efficient energy transfer would be provided then alkali-earth fluorides are perspective scintillators. Real light yield of CaF_2–Eu is 18000–22000 ph/MeV, but BaF_2 and BaF_2–Ce demonstrate low light yield at about 10000 ph/MeV [4]. Scintillation properties of SrF_2 crystals almost are not investigated. Light yield of SrF_2 is estimated about 10000–12000 ph/MeV in Ref. [5]. However, potential light yield of SrF_2 can be higher. Also SrF_2 crystals doped with Ce^{3+} and Pr^{3+} have a temperature stability of light yield in wide range (–50 to 200 °C) [6]. Therefore, SrF_2 crystals can be perspective scintillator for well-logging. So scintillator properties of strontium fluoride crystals are among the least studied of fluorides crystals, but these crystals have a potential application. Thus, the investigations of scintillation properties of strontium fluorides are topical today.

2. Experimental methodology

The crystals pure SrF_2 and doped with different concentrations of Ce^{3+} ions were grown in a graphite crucible by the Stockbarger method. We used several experimental techniques in measurement of scintillation properties of the crystals. To determine light yield of the crystals pulsed-height spectra under ^{137}Cs 662 KeV gamma source excitation are measured. To record the spectra PMT FEU-39A, a homemade preamplifier and an Ortec 570A spectrometric amplifier are used. The crystal was covered with several layers of ultraviolet reflecting Teflon tape (PFTE tape). The shaping time of Ortec 570 spectrometric amplifier was set at 10 μs to collect much light from scintillator.

Also X-ray emission spectra are studied. The excitation of the emission was performed with X-ray tube with Pd anode, operational voltage was 35 kV, and current was 0,8 mA. For registration PMT FEU-39A, grating monochromator VM-4 was used. The spectra recorded at photon-counting regime.

Decay curves measured under pulsed X-ray excitation performing pulsed X-ray tube MIRA (100 KeV, pulse duration about 8 ns), and ^{137}Cs E = 662 KeV gamma source. The curves were registered by oscilloscope Rigol DS-1202CA.

3. Experimental results and discussion

Figure 1 shows spectra of X-ray luminescence of pure SrF_2, NaI–Tl, SrF_2 – 0,3 mol. % Ce^{3+}, and CaF_2 – 0,1 mol. % Eu^{2+}. In the spectrum of SrF_2 a wide band at 280 nm is attributed to self-trapped exciton (STE) emission. In SrF_2 doped with Ce^{3+} ions STE luminescence is quenched and vanished at con-

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centrations Ce^{3+} ions higher than 1 mol. %. The most intense bands in X-ray luminescence spectra of SrF_2-Ce^{3+} crystal at 310 and 325 nm correspond to 5*d*–4*f* emission of Ce^{3+} ions. Light output of the samples can be estimated by areas under emission curves. In the inset of Fig. 1 concentration dependence of light output of SrF_2–Ce^{3+} crystals normalized to light yield of pure SrF_2 is presented. The highest light yield was found in the crystal doped with 0,3 mol. % of Ce^{3+} ions.

Light output of X-ray luminescence of SrF_2 and SrF_2–Ce^{3+} crystals is compared with the luminescence output of NaI–Tl crystal. Results are shown in Table I. Light yield of NaI–Tl crystals is about 43000 ph/MeV, therefore light yield of the measured samples can be estimated. The data are presented in Table 1. The value of light yield of CaF_2 – 0,1 mol. % Eu^{2+} is about 21500 ph/MeV that is in according with known data for CaF_2–Eu crystals [3]. For pure SrF_2 light yield is about 20640 ph/MeV, for SrF_2 doped with 0,3 mol. % crystals light yield is about 34000 ph/MeV, for doped with 1 mol. % crystals – 18500 ph/MeV.

Figure 2 shows pulse height spectra of SrF_2, SrF_2 – 0,3 mol. % Ce^{3+} and NaI–Tl. Energy resolution of photopeak of NaI–Tl is about 6,7 %, SrF_2 – 10 %, SrF_2 – 0,3 mol. % Ce^{3+} – 9,3 %. Light yield of pure SrF_2 crystal is about 42 % of NaI–Tl that is similar to x-ray emission light output (see Table 1). Light yield of SrF_2 crystals doped with 0,3 mol. % Ce^{3+} ions is about 32 % of NaI–Tl that is less than X-ray luminescence light output. These data are collected in Table I. It should be noted, that in Table I all data are not corrected for spectral sensitivity of registration channel. It is well-known [7] that spectral sensitivity of S20 photocathode (PMT FEU 39A) is higher at 400 nm than at 280–330 nm.

Scintillation decay time profile of SrF_2 – 0,3 mol. % Ce^{3+} is shown in Fig. 3. Resistance of oscilloscope input is selected equal 2,6 KΩ for registration long time decay components in Ce^{3+} emission. First components (2,8 μs) in Ce^{3+} decay is integrated short component. Lifetime of this component is equal 130 ns at 50 Ω input resistance. Contribution of slow components to the total luminescence can be up to 50 % in SrF_2 – 0,3 mol. % Ce^{3+}.

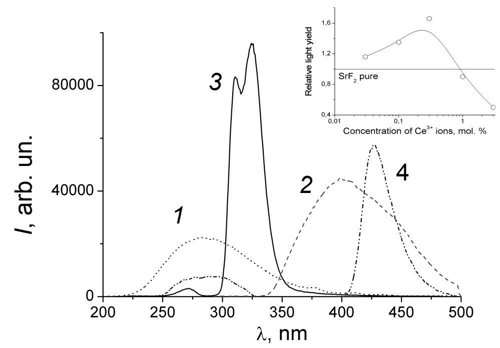

Fig. 1. Luminescence spectra of SrF_2 (curve *1*), NaI–Tl (curve *2*), SrF_2 – 0,3 mol. % Ce^{3+} и CaF_2 – 0,1 mol. % Eu^{2+} under X-ray excitation. In inset concentration dependence of light yield of SrF_2–Ce^{3+} crystals is presented. All light yields were normalized to light output of pure SrF_2 crystals.

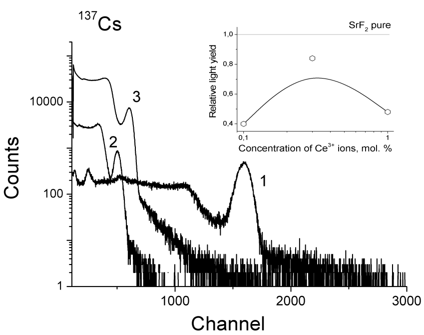

Fig. 2. Pulse height spectra of NaI–Tl (curve *1*), SrF_2 – 0,3 mol. % Ce^{3+} (curve *2*) and SrF_2 (curve *3*) under gamma-source ^{137}Cs (*E* = 662 keV) excitation. In inset concentration dependence of SrF_2–Ce^{3+} light yield measured by photopeak position is presented. Light yield of pure SrF_2 is equal 1.

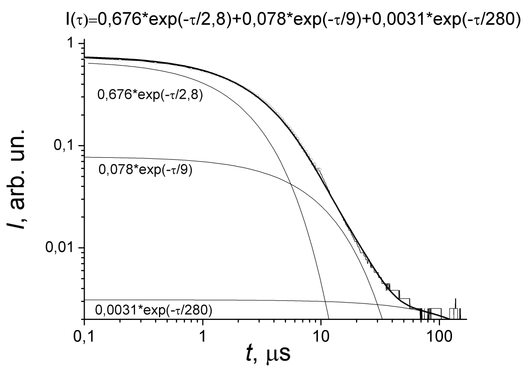

Fig. 3. Scintillation decay time profile of SrF_2 – 0,3 mol. %Ce^{3+} crystal measured under gamma-source ^{137}Cs excitation (*E* = 662 keV). Exponential components of total decay curve are shown separately.

Table 1

Light outputs of SrF_2, SrF_2–Ce^{3+}, NaI–Tl и CaF_2–0,1 mol.% Eu^{2+} crystals measured under X-ray and gamma excitation

Scintillator	Light output measured from X-ray luminescence spectra		Light output measured from pulse height spectra	
	rel. un.	ph/MeV	rel. un.	ph/MeV
NaI–Tl	1	43000	1	43000
CaF_2 – 0,1 mol. % Eu	0,5	21500	0,44	18920
SrF_2	0,48	20640	0,42	18060
SrF_2 – 0,3 mol. % Ce^{3+}	0,79	33970	0,32	13760
SrF_2 – 1 mol. % Ce^{3+}	0,43	18490	0,2	8600

Therefore, the difference in light yields measured from x-ray luminescence and pulse height spectra is explained by presence of intensive slow components in cerium ions luminescence (see Fig. 3). They give a large contribution to total light output. Shaping time of pulse height spectrum measurement is 10 µs, and a large part of emitted light is not registered. Shaping time of X-ray luminescence spectra is higher. It is amount about 1 s.

Working knowledge is as follows. Estimations of temperature stability of light yield [6] are pointed that crystals of SrF_2 – 0,3 mol. % Ce^{3+} и SrF_2 – 1 mol. % Ce^{3+} demonstrate high temperature stability of light output in the region between –50 and 200 °C. For this reason, the SrF_2–Ce^{3+} crystals would be perspective scintillators for well-logging applications.

The SrF_2 crystals have a higher than NaI–Tl density – 4,18 g/cm^3 and comparable light yield, and it is no hygroscopic. Light yield of SrF_2–Ce^{3+} samples can be increased by decreasing of slow component contribution in Ce^{3+} luminescence. It might be possible by co-doping these crystals with Ga^{3+} or In^{3+} ions. In future, uncorrected light yield of SrF_2–Ce^{3+} crystals may be risen up to 34000 ph/MeV.

4. Conclusion

Crystals SrF_2 – 0,3 mol. % Ce^{3+} и SrF_2 – 1 mol. % Ce^{3+} have a high light yield, no hygroscopic, temperature stability of light output. Therefore, these crystals are perspective scintillators for well-logging applications.

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REFERENCES

1. Dorenbos P., IEEE Trans. on Nucl. Sci., **57**, 1162–1167 (2010).
2. Alekhin M.S. et al., IEEE Trans. Nucl. Sci., **58**, 2519–2527 (2011).
3. Derenzo S.E., Database: http://scintillator.lbl.gov.
4. Visser R., et al., IEEE Trans. Nucl. Sci., **38**, 178–183 (1991).
5. Schotanus P. et al., IEEE Trans. Nucl. Sci., 34, 272 (1987).
6. Shendrik R., Radzhabov E., IEEE Trans. Nucl. Sci., **57**, 1295–1299 (2010).
7. Flyckt S.O., Photomultipliers tubes. Photonis, France (2002).

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