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New Radiation Resistant Scintillator LFS-3 for Electromagnetic Calorimeters

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Abstract—The results of the study of optical and luminescence characteristics of new LFS-3 heavy scintillation crystals are presented. Advantages of these crystals in comparison with conventional scintillators are discussed. The radiation resistance of LFS-3 scintillation crystals is studied using an intense ^{60}Co radioactive source and a proton beam with an energy of 155 MeV. No changes in the optical transmission of LFS-3 crystals after their irradiation with a dose of 23 Mrad are detected.

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High beam energies and luminosities of modern accelerator systems impose increased requirements for radiation resistance of materials used in detectors of experimental setups. The development of new high-density scintillator crystals which can be used under conditions of high radiation loads is urgent for both the electromagnetic calorimeter of the Compact Muon Solenoid (CMS), operating at the CERN, and for calorimetry at new linear colliders (ILC and CLIC) whose development is planned in the nearest years [1]. Since radiators 20–25 radiation length long are used in experiments of high-energy physics, even a slight decrease in the optical transmittance per unit length, caused by radiation damages, can result in a significant degradation of electromagnetic calorimeter characteristics. Recently, it was found that modules of end-face parts of the CMS electromagnetic calorimeter, developed based on $PbWO_4$ crystals, are damaged by high-energy hadrons during the LHC collider operation. For this reason, it is planned to replace $PbWO_4$ crystals by the next-generation radiation resistant crystals.

One of the promising scintillators are crystals based on lutetium orthosilicate. LSO ($Ce_xLu_{2-x}SiO_5$) scintillation crystals were proposed and studied by C. L. Melcher and J. S. Schweitzer in 1992 as new materials to be used in gamma detectors [2]. LYSO ($Ce_x(Lu, Y)_{2-x}SiO_5$) crystals were first grown and studied by D. W. Cooke in 2000 [3].

Large LSO and LYSO crystals were proposed for using in experiments of high-energy physics as promising materials for homogeneous electromagnetic calorimeters [4]. The first prototype of the electromagnetic calorimeter based on 3×3 LYSO crystals was successfully tested at the MAMI accelerator in the photon energy region to 490 MeV [5].

The radiation resistance of LSO and LYSO crystals during their irradiation with a γ-ray beam was studied in [6, 7]. It was noted that a decrease in the optical transmittance of the LSO crystal was $\sim$2.5% per centimeter under irradiation with a dose of 10 Mrad from a ^{60}Co radioactive source [6].

A disadvantage of LSO crystals is a large parameter spread when fabricating samples from the same initial crystal. LYSO crystals have a similar nonuniformity of scintillation parameters along the boule length and tend to cracking.

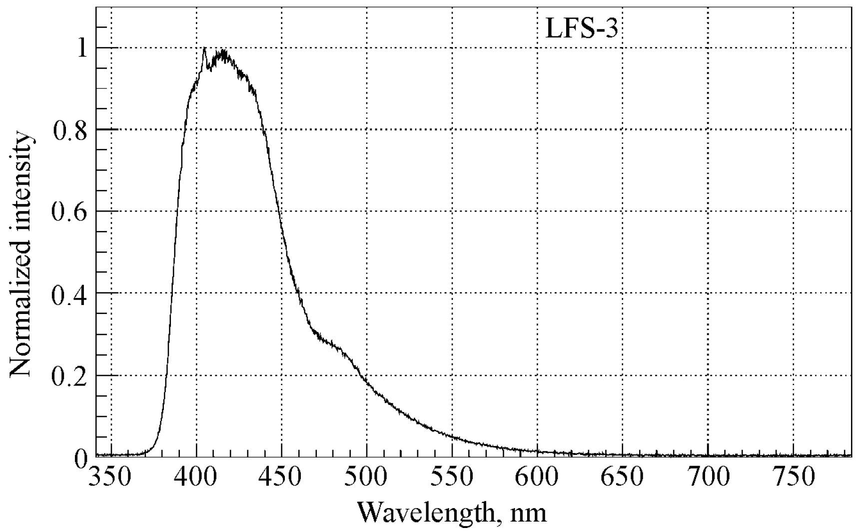

Fig. 1. X-ray luminescence spectrum for the LFS-3 crystal.

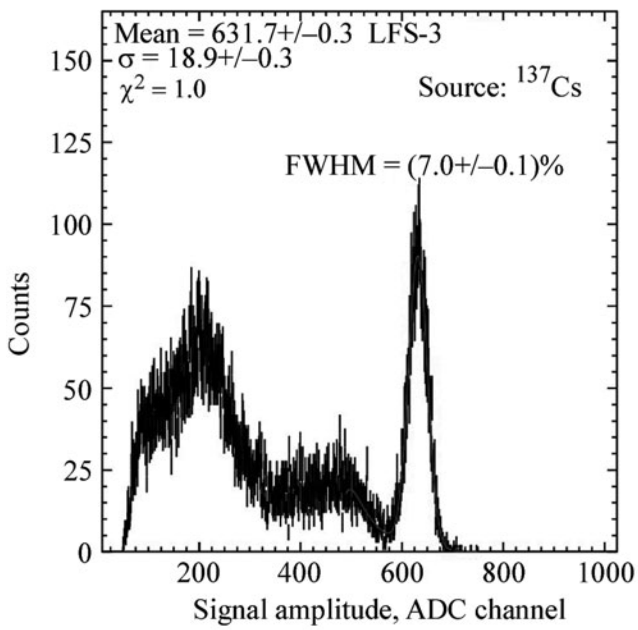

Fig. 2. Amplitude distribution of the LFS-3 scintillator irradiated with γ-rays of the ^{137}Cs source.

New LFS-3 (Lutetium Fine Silicate) scintillators were developed at the Prokhorov General Physics Institute, based on lutetium silicate crystals which crystallize into a monoclinic system with space group $C2/c$, $Z = 4$. The composition LFS-3 $\mathrm{Ce}_x\mathrm{Lu}_{2+2y-x-z}\mathrm{A}_z\mathrm{Si}_{1-y}\mathrm{O}_{5+y}$ was patented, where A is at least one element chosen from the group containing Ca, Gd, Sc, Y, La, Eu, and Tb [8].

LFS-3 scintillation crystals 90 mm in diameter and 200 mm long were grown by the Zecotec Imaging Systems Pte Ltd. using the Czochralski method. The initial melt was prepared using Lu_2O_3, SiO_2 components and a CeO_2 scintillation additive with a purity of 99.99%. From initial boules, LFS-3 crystal samples were cut out for studying their optical and luminescence characteristics.

The emission spectrum of LFS-3 crystals (Fig. 1) was measured using a setup with luminescence excitation by X-ray photons with an energy of 30 KeV.

To determine the light yield of LFS-3 scintillators, the total-absorption spectra of γ-rays from radioactive sources, i.e., the so-called photopeaks, were used. In this study, the scintillator photopeaks

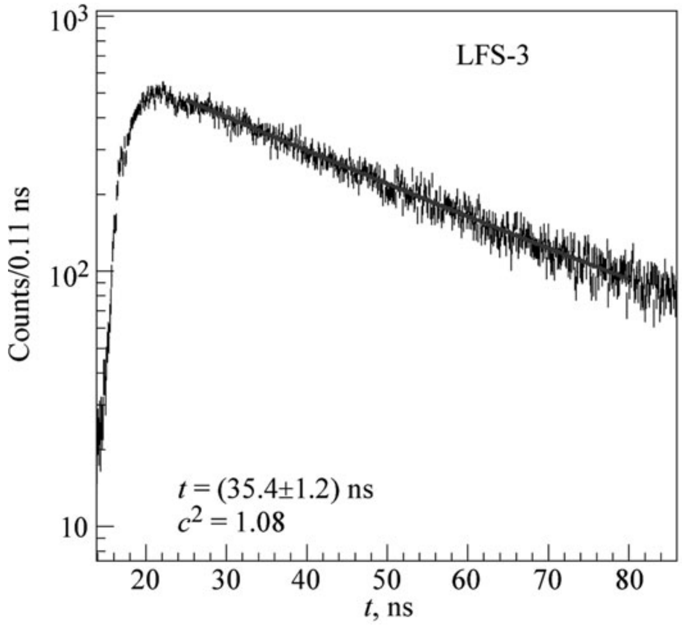

Fig. 3. Time dependence of the LFS-3 scintillator luminescence intensity.

were measured with the setup consisting of a Hamamatsu R4125Q photomultiplier with a quartz window, an ORTEC 579 fast amplifier, and an ADC LeCroy 2249W charge-sensitive amplitude converter. Figure 2 shows the photopeak for the LFS-3 crystal in which scintillations were excited by γ-rays of the ^{137}Cs radioactive source.

The absolute light yield in the units of the number of photoelectrons per MeV was determined using the calibration of amplitude converter (ADC) channels by the single-electron peak position. Using the average quantum efficiency of the R4125Q photomultiplier photocathode in the region of the LFS-3 emission spectrum to convert the number of photoelectrons per MeV to the photon yield, we obtain the LFS-3 light yield of 38000 photon/MeV.

The luminescence time of LFS-3 scintillators was studied with a special installation using the delayed coincidence method consisting in measurements of the distribution of time intervals Δt between scintillator excitation and photoelectron formation on the photomultiplier photocathode with the aid of a time-to-digital converter (TDC). Figure 3 shows the time dependence of the LFS-3 crystal luminescence intensity, obtained using a ^{22}Na radioactive source. The parameters of the time spectra were determined using the function

$$I(t) = \sum A_i \cdot \exp(-t/\tau_i) + C,$$

where τ_i is the ith scintillator luminescence time component, A_i is its amplitude, and C is the accidental coincidence background.

The LFS-3 crystal luminescence time is 35.4 ± 1.4 ns for fitting the time spectrum by a one-exponent function.

The main characteristics of new LFS-3 crystals in comparison with NaI(Tl) and LYSO crystals are given in Table 1. It should be noted that LFS-3 is a promising scintillation material for both the electromagnetic calorimetry in high-energy physics experiments and the positron-emission tomography.

In this work, the stability of LFS-3 crystals to radiation damages caused by charged hadrons and γ-rays was studied.

The radiation resistance of LFS-3 crystals was measured using polished samples $10 \times 10 \times 10$ mm^3 in size cut out from various regions of the initial boule. The samples were irradiated by the ^{60}Co radioactive source (the maximum power is $\sim$4 krad/min). LFS-3 crystals were sequentially irradiated with three doses: 5, 23, and 68 Mrad. The optical transmission spectra were measured before and

Table 1. Main characteristics of scintillation crystals

Material	NaI(Tl)	LYSO [9]	LFS-3
Density, ρ g/cm^3	3.67	7.1	7.35
Melting point, °C	651	1990	2000
Radiation length, X_0, cm	2.59	1.19	1.15
Moliere radius R_m, cm	4.3	2.16	2.09
Light yield (photon/MeV)	40000	32000	38000
Luminescence time, ns	230	41	35
Emission maximum, nm	410	420	425
Refractive index n at the emission maximum	1.85	1.81	1.81
Hardness, Mohs	2	5	5
Hygroscopicity	Yes	No	No

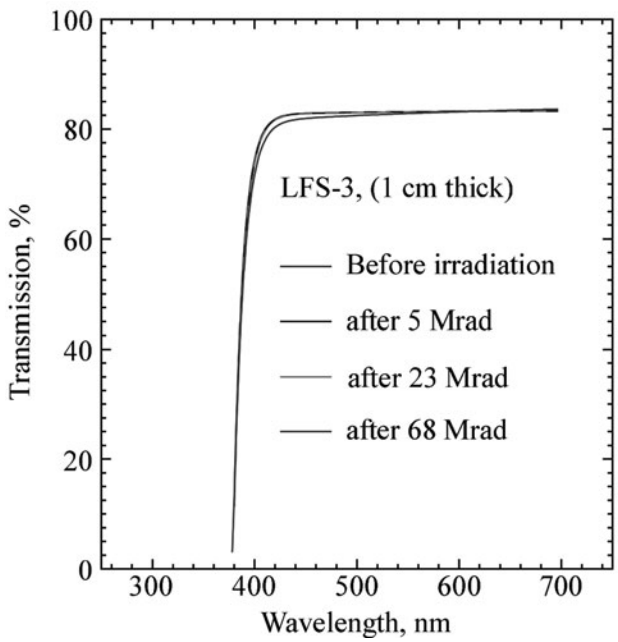

Fig. 4. Optical transmission spectra for LFS-3 crystals before and after irradiation with γ-rays of the ^{60}Co radioactive source.

immediately after irradiation using a Kruess Optronic VIS 6500 spectrophotometer. The results for one of the LFS-3 crystals are shown in Fig. 4. An analysis of the transmission spectra shows that the optical transmittance in the LFS-3 emission region decreases by ~2.5% for a dose of 68 Mrad. For the LFS-3 samples cut out from the upper, middle, and lower parts of the initial crystal, the dose of 23 Mrad had no appreciable effect on optical transmission [10].

It is known that irradiation of crystals with high-energy hadrons can damage the crystal lattice and generate a significant number of defects. Such damages can not only decrease the optical transmittance of crystals, but also disturb the crystal scintillation mechanism.

Radiation damages of LFS-3 crystals during their irradiation with hadrons was studied in the proton beam of the synchrotron of the Institute of Theoretical and Experimental Physics (ITEP). For

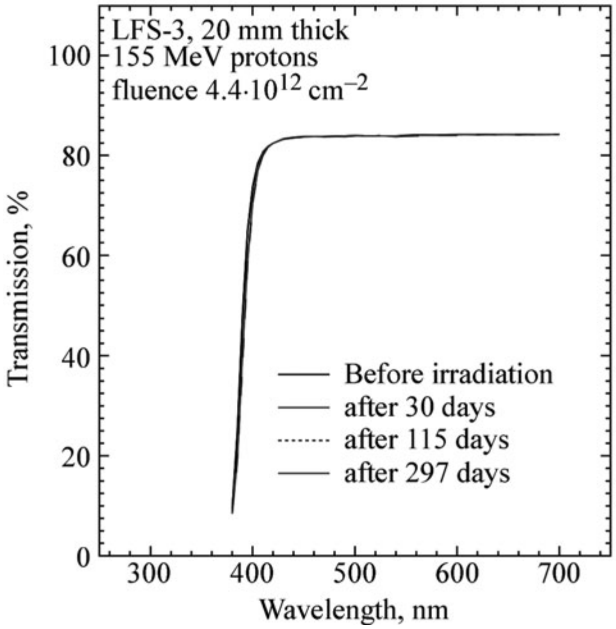

Fig. 5. Optical transmission spectra of LFS-3 crystals before and after irradiation with the 155-MeV proton beam.

the irradiation in the proton beam, polished LFS-3 samples 20 mm long and 11×11 mm^2 in the cross section were used. LFS crystals were packed into a 3×2 matrix for simultaneous irradiation of six samples in the 50-mm-diameter proton beam. All LFS crystals were irradiated with 155-MeV protons to a fluence of $4.4 \cdot 10^{12}$ particle/cm^2. Due to the high level of induced radioactivity, the first measurements of the optical transmittance of crystal samples irradiated with protons were performed only in 30 days after irradiation. The optical transmission spectra for LFS-3 crystals before and after irradiation with the proton beam are shown in Fig. 5. We note that there are no damages in LFS-3 crystals for a fluence of $4.4 \cdot 10^{12}$ particle/cm^2 [11].

The results obtained show that the new LFS-3 high-density scintillator crystal is the best among crystalline materials intended for electromagnetic calorimeters capable of operating under conditions of high radiation loads.

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