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Radiation damage effects in Y_2SiO_5:Ce scintillation crystals under γ-quanta and 24 GeV protons

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ABSTRACT

This work focuses on the study of changes in the optical transmission of Y_2SiO_5:Ce crystals caused by ionizing radiation from γ-quanta and high energy protons. Radioisotope content of proton-irradiated crystals, transmission and induced absorption spectra, and scintillation characteristics are measured after irradiation with protons. In contrast to crystals of heavy complex oxides, Y_2SiO_5:Ce crystals do not demonstrate significant deterioration of transmission in the luminescence range (400–600 nm) under irradiation. Such crystals can be considered as a material for construction of detecting cells of the calorimetric detectors at LHC with high luminosity. The feasibility of growing large crackless Y_2SiO_5:Ce crystals with a diameter up to 50 mm and length up to 250 mm is demonstrated.

1. Introduction

Heavy and fast crystalline scintillators are widely used in calorimeters for current and future experiments in high energy physics. For example, lead tungstate, $PbWO_4$ (PWO), is used in the electromagnetic calorimeter in CMS (ECAL) and the photon detector ALICE at LHC (CERN) [1,2]. PWO crystals demonstrate good radiation hardness under γ-irradiation. High-quality PWO crystals have provided a stable process of data accumulation and acquisition in CMS ECAL in LHC radiation environment from the start of its operation. Further research program at LHC requires a significant increase in the accelerator luminosity up to 500 fbn^{-1}in a each year from 2021. At such luminosity charged hadrons, which after 2021 will be capable of producing an integral flux with a density reaching 10^{14} particles/cm^2, become an important damaging component of ionizing radiation near the particle collision area. Thus, the selection of scintillation materials with moderate radiation damage at irradiation by high-energy hadrons is the basic task in preparing a scientific background of LHC detector setup modernization. A fast and bright scintillation material with orthosilicate structureY_2SiO_5:Ce (YSO:Ce) seems to be a promising candidate. It contains light elements with a relatively small cross-section of nuclear reactions under high-energy hadrons. This work studies optical transmission damage of YSO:Ce crystals by different components of ionizing radiation. The crystals were irradiated with a γ-quanta ^{60}Co source with the absorbed dose 2000 Gy and a high-intensity flux of 24 GeV protons at CERN PS with the integral flux of $\sim 3.6 \times 10^{13}$ particles/cm^2. The crystals demonstrated small changes in optical transmission in the luminescence band, because the majority of color centers are manifested in the spectral range near the fundamental absorption edge. We discuss the mechanism of radiation damage of YSO:Ce crystals from the analysis of radioisotope content after irradiation with high-energy protons. We also show a feasibility of growing a high-quality large YSO:Ce crystal by the Czochralski method.

2. Growth of large YSO:Ce scintillation crystals

To perform this study, a series of large crack-free YSO:Ce crystals with a diameter up to 50 mm and length up to 250 mm was grown by the Czochralski method in ISMA (Fig. 1). We used 4 N purity chemicals to prepare the raw material for the crystal growth. Crystals were pulled on a seed from the melt using Ir crucibles with a diameter and a length of 90 mm in Ar atmosphere. The crystal diameter was controlled by a weight sensor. The crystal growth rate was 1.5–2 mm/h; the crystal rotation rate was 30–35 rot/min. The basic complications in growing large YSO:Ce crystals comprise cracking of the grown ingots and deformation of Ir crucibles. Crystal cracking is attributed to high thermal gradients above the crucible. Those were eliminated using an additional independent heater

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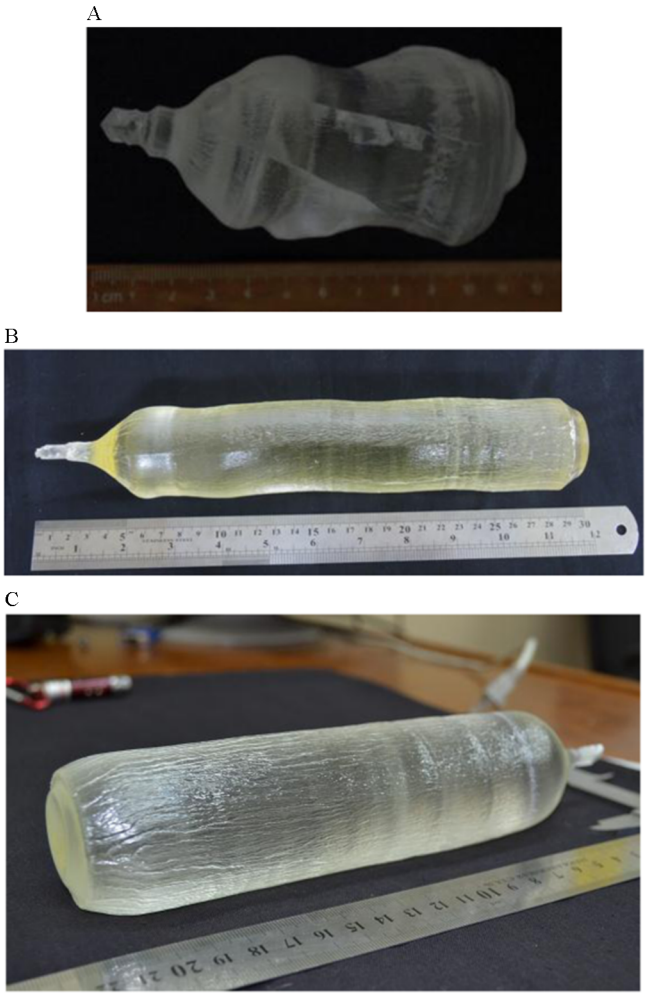

Fig. 1. Large-size YSO:Ce single crystals grown in ISMA by the Czochralski method: as grown before (A) and after (B) optimization of heating conditions, grown in optimized conditions and annealed in oxidizing atmosphere (C). (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

installed above the crucible and providing a flatter axial thermal gradient. Crucible deformation is caused by the crystallization of melt remnants inside the crucible after the crystal growth. Different heat expansion coefficients of the crucible and crystallized melt lead to an expansion of the crucible in its lower part. The crucible deformation was reduced by minimizing the melt volume remaining inside the crucible after the crystal cut-off. Optimization of thermal conditions at the crystallization interface provided the crystallization of 80–90% of melt in the crucible. The grown ingots had yellow tint (Fig. 1B), which was eliminated (Fig. 1C) by annealing in oxidizing atmosphere in accordance with the procedure described in [3]. Post-growth annealing was also necessary to prevent crack formation in crystals during cutting and polishing.

Since the raw material price mainly determines the crystal cost at large scale production, YSO:Ce is dramatically cheaper compared to its well-known ortho-silicate analogs—Lu_2SiO_5:Ce, $Lu_{2-2x}Y_{2x}SiO_5$:Ce, and Gd_2SiO_5:Ce. Yttrium oxide is 10–100 times cheaper than lutetium oxide. Therefore YSO:Ce is an efficient and relatively cheap scintillation material for particle physics experiments requiring a huge amount of detecting material. High cost of Ir is a disadvantage of YSO:Ce, as well as many other high-melting-point crystals; however, the method used to prevent the crucible from deformation sufficiently increases its lifetime.

The homogeneity of Ce axial distribution in YSO:Ce should be better than that in LSO:Ce or LYSO:Ce, because the activator distribution coefficient k_{eff} at Czochralksi growth of rare-earth orthosilicates increases as the difference between the host and activator ionic radii is decreased [4]. For this reason, the values of k_{eff} reported for YSO:Ce were 0.25 [4] and 0.34 [5], while those reported for LSO:Ce were 0.18–0.22 [6–8]. The Ce radial distribution is assumed to be negligible, because under the optimized conditions of Czochralksi growth (see Fig. 1C), almost flat crystallization interface is sustained.

3. Radiation-induced damage in light and heavy crystalline materials

To characterize the radiation damage in crystals, reference samples with the volume 1 cm^3 were cut from the middle part (Samples 1 and 2) of the ingots. All measurements were performed at room temperature. Optical transmission of samples was measured before and after irradiation. Varian Spectrophotometers Cary 1E and Cary50 were used for measurements. The amplitude spectra and scintillation kinetics were measured with a custom-made spectrometer.

The samples were studied using the procedure recently applied to studying LSO:Ce radiation hardness [9]. First, they were irradiated using a ^{60}Co source (1.22 MeV, 2000 Gy absorbed dose, dose rate 1000 Gy/h). Then the samples were irradiated with a 24 GeV/c proton beam at the CERN Proton Synchrotron (PS) with a flux of about 10^9 p/cm^2/s, to the fluence of about 3.6×10^{13} p/cm^2, estimated by the Al-foil radiation monitor. 10 cm long $PbWO_4$ (PWO) crystal was used in irradiation unit to produce cascade of secondary particles. Samples were mounted at the front and rear parts of a 10-cm-long PWO crystal to incident proton beam, correspondingly. It allowed to study a possible difference in the damage produced by pure incident protons (sample in front of the beam) and that produced by the mixture of the protons and the cascade of secondary particles (sample after the PWO crystal).

The transmission of samples irradiated by gamma quanta has been measured 30 min after γ-irradiation. After irradiation with protons samples were kept at zone of irradiation until level of induced radioactivity allowed their safe transportation to the protected store. They were under permanent control of the induced radioactivity in the store to define their ability for measurements. The optical transmission spectra and set of radioisotopes produced in the samples by protons were measured 30 days after proton irradiation, as soon as the induced radioactivity of the samples dropped to the level safe for personnel. Irradiation and manipulations with irradiated crystals were performed by experts of appropriate CERN Service.

Radiation damage effects caused by γ-irradiation depend on the initial perfection of the crystal [10]. This means that irradiation with γ-quanta with the energy below 5 MeV, even with a high dose rate up to 10^6 Gy/h, does not create new defects in a scintillation material due to a weak interaction of quanta with nuclei. Charged particles create additional defects in crystals. Heavier charged particles, such as protons, hadrons, or nuclear fragments, lose much more energy and produce a shower of knocked-out ions in the crystal. For this reason, the damage of crystals under high-energy charged particles is a more complicated phenomenon. To date, there has been progress only in understanding the mechanism of PWO, LSO, and YSO scintillator damage under proton irradiation [9,11–13]. It was assumed that the damage of a crystalline material produced by high-energy hadrons passing through the crystal is caused by a combination of effects from primary particles and from light fragments of nuclear interactions. Two types of hadron-nucleus reactions are initiated in heavy crystals: spallation and fusion. A set of radioisotopes, which is created in the crystal under high-energy protons,

Table 1
Measured radio-isotopes in YSO:Ce crystal after irradiation with protons with the fluence 3.6×10^{13} p/cm^2.

Isotope	Isotope half-life, days	Isotope activity, Bq/cm^3 ($E\gamma \geq 100$ keV), 30 days after irradiation	Concentration of atoms of irradiation-created isotopes, 1/cm^3
Rb-83	8.62E+01	2220	3.04E+10
Rb-84	3.20E+01	3.60E+02	2.75E+09
Y-88	1.07E+02	6700	1.09E+11
Zr-88	8.34E+01	1.30E+02	1.73E+09

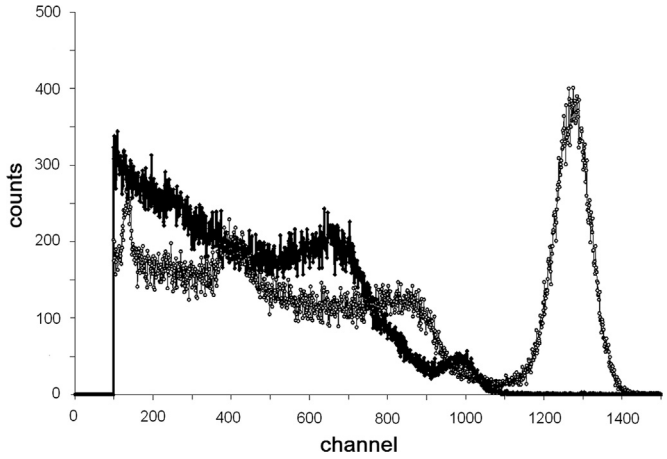

Fig. 2. Pulse height spectra of ^{137}Cs (662 keV) source measured at room temperature with the sample 1 (YSO:Ce, filled dots) compared to NaI(Tl) sample with the dimensions 1" × 1" (empty points).

enables one to find out which reaction mechanism dominates in the crystals [9].

The set of radioisotopes detected in the irradiated YSO samples is given in Table 1. The presence of long-living radioisotopes with a mass close to that of Y in the irradiated crystal indicates that the reactions with creation of the lightest fragments, such as deutron (d), triton (t), and He nuclei, are dominating. This fact is perfectly consistent with the suggestion that spallation [9,14] is the dominating interaction mechanism of high energy protons with atoms of the crystal.

4. Optical and scintillation properties

YSO:Ce is a bright and fast scintillation material. Fig. 2 shows amplitude spectra measured for Sample 1 using ^{137}Cs (662 keV) source with the reference NaI(Tl) crystal of 1′ × 1′ dimensions. Its light yield (LY) is close to 75% of NaI(Tl) at room temperature and it possesses a single exponential kinetics of scintillation with a decay time of 60 ns, as seen in Fig. 3.

Fig. 4(a and b) shows the optical transmission spectra of Sample 1 before and after γ-irradiation and the induced absorption spectrum, correspondingly. The spectrum measured with Sample 2 is identical to that measured with Sample 1 after similar γ-irradiation. The crystal contains 0.2 at% of activating ions having strong absorption in UV range due to 5f→4d electron transitions. So a large part of the transmission spectrum from the crystal fundamental absorption cutoff (200 nm) to the long wavelength shoulder of the lowest absorption transition (370 nm) of activator is therefore out of observation. Nevertheless, for crystal application in irradiation environment, the most important part of the spectrum is 370–600 nm in which the scintillation band is located.

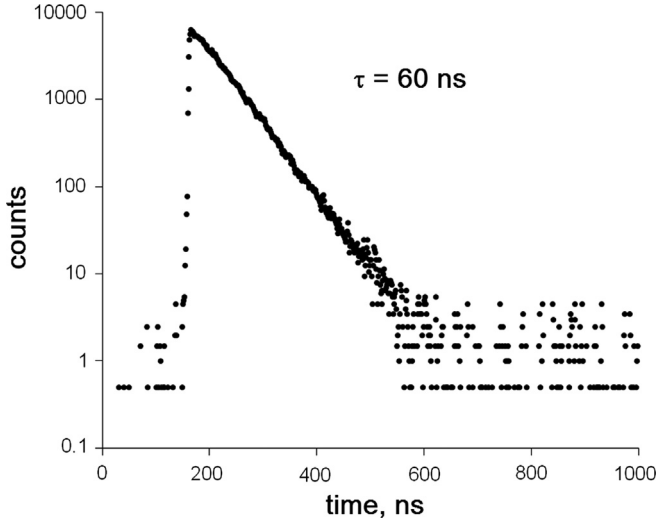

Fig. 3. YSO:Ce scintillation decay measured at room temperature.

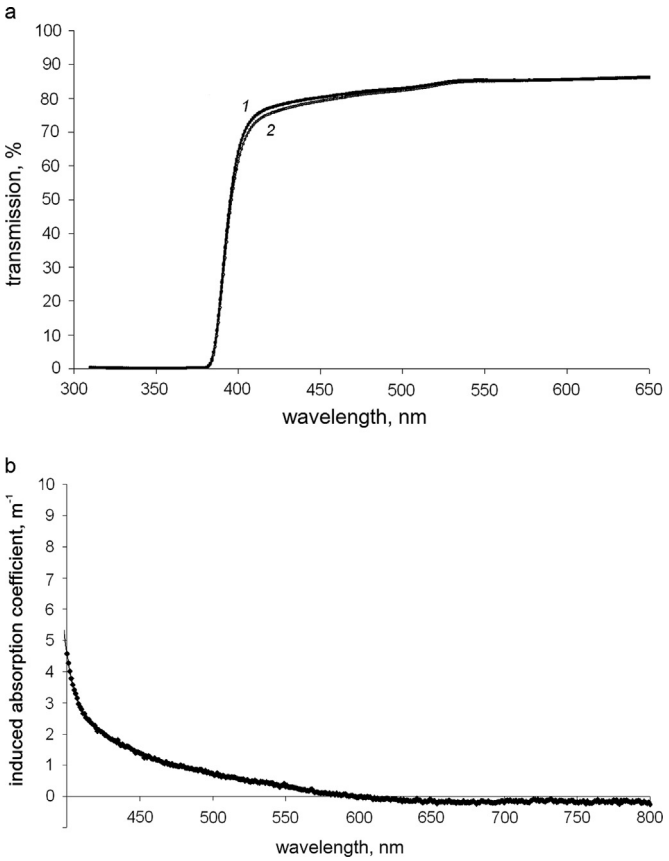

Fig. 4. (a) Transmission spectra of YSO:Ce (Sample 1) before (1) and after (2) irradiation by ^{60}Co source (1.22 MeV, absorbed dose 2000 Gy, irradiation intensity — 1000 Gy/h). (b) Induced absorption spectrum.

A minor shift of the sample induced absorption curve below zero is observed in the spectral range $\lambda > 600$ nm. Most probably, it occurs due to slightly different disposition of the crystal in the spectrometer sample unit before and after irradiation.

The optical transmission spectra of Sample 2 before and after irradiation with protons are shown in Fig. 5. It is interesting to note that Sample 2, which was mounted after PWO crystal during irradiation, showed even smaller deterioration of optical transmission than Sample 1. Similar to the case of γ-irradiation, no strong deterioration of optical transmission was observed in the actual

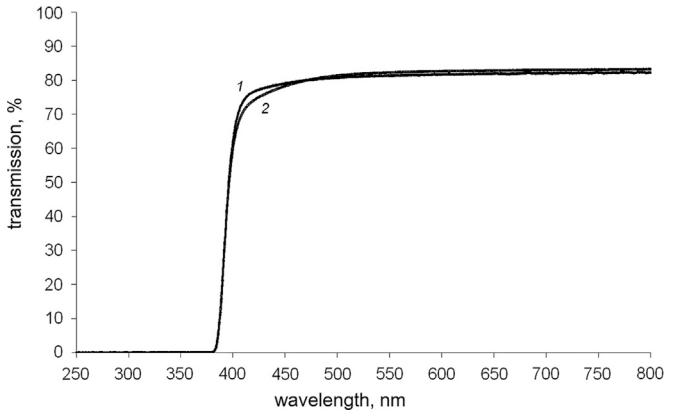

Fig. 5. YSO:Ce (Sample 2) transmission measured before irradiation by the 24 GeV proton beam (1) and 30 days after irradiation (2).

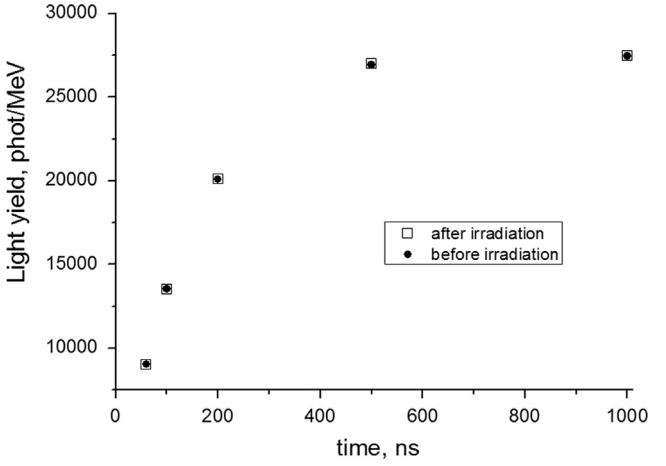

Fig. 6. Light yield of Sample 1: irradiated with protons (black dots) and after subsequent irradiation by γ-quanta with the absorbed dose 100 Gy (empty squares). Measurement after γ-quanta irradiation was started 2 min after irradiation.

spectral range after irradiation with protons. Light yield of both samples dropped by less than 10% after irradiation with γ-quanta, and a slightly greater drop of about 12% was observed one month after irradiation with protons.

The obtained data show that there is no significant difference in the crystal induced absorption spectra under different kinds of irradiation. This indicates that in a light host like YSO an elastic interaction of primary particles and secondary particles with nuclei of the matrix does not produce a large number of defects. Most probably, the concentration of new defects is comparable or even less than that of post-growth defects.

Moreover, irradiation does not affect Ce^{3+} concentration in the crystal. The amplitude of the absorption band or its intensity at a certain wavelength at the long wavelength shoulder is proportional to the concentration of the activator in the trivalent state. Neither sample showed a change in the spectrum cutoff indicating stability of the Ce^{3+} ions in the crystal. A possible mechanism of the charge state change $Ce^{3+} \rightarrow Ce^{4+}$ is a direct ionization of Ce^{3+} ions by the incident ionizing radiation. A fraction of electrons after the ionization of Ce^{3+} ions is captured by the nearby oxygen vacancies. Since the concentration of vacancies is small, the probability of electron capture is not high. Perfect YSO:Ce crystals were found to be free from the recharge processes at the achieved level of particle fluence. This correlates with the fact that no sufficient difference in the crystal light yield after different irradiations is observed. The data on LY measured with different gates (Fig. 6) also support this suggestion. No change in the gated LY was observed in Sample 1 irradiated with protons and with a subsequent irradiation by γ-quanta with the absorbed dose 100 Gy. The similarity of LY at different gates indicates a small level of the crystal phosphorescence which occurs in Ce-doped crystals due to a large concentration of oxygen vacancies.

5. Conclusions

Modification of scintillation parameters and induced absorption of Ce-doped yttrium silicate crystals under γ- and 24 GeV proton irradiation has been investigated. No significant difference was observed in the damage effects produced by both irradiations in a Ce-doped YSO crystal. This indicates that high-energy protons do not produce a sufficient concentration of the defects and dedicated color centers in the crystal to deteriorate significantly its scintillation properties.

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