

Stability of Spectrometric Characteristics of CsI:Tl Detectors Depending on the Surface Treatment Method

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Abstract—The influence of various mechanochemical methods used to treat the entrance surface of a CsI:Tl scintillator on the specific light output and the pulse-height resolution during detection of α particles with energies of 2 or 5 MeV is described. Nonlinearity between the light output and the energy of α particles increases from 7% for a polished surface to 35% after grinding, which leads to a degraded pulse-height resolution. A method for treating the CsI:Tl surface is proposed that minimizes the light-output nonlinearity and ensures stability of spectrometric characteristics in time.

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INTRODUCTION

Stability of spectrometric characteristics is an important performance parameter of scintillators, especially when detecting weakly penetrating radiations (α and β - particles and X rays). It is known that the state of the scintillator surface [1] and, consequently, the technique used for its preparation [2] have a considerable effect on both the characteristics themselves (the light output and the energy resolution) and their stability during operation. Depending on their functions, the surfaces of the detector's sensitive element can be divided into the following:

(i) the entrance surface for the incident radiation, in the subsurface layer of which short-range radiation interacts with the scintillator substance;

(ii) the lateral surface through which scintillation light leaves the scintillator volume, in particular to reflectors; and

(iii) the output surface through which scintillation light leaves the crystal and falls on a photoreceiver [2].

The methods for treating these functionally different scintillator surfaces have specific features. The subsurface layer of the entrance surface must contain a minimum number of defects, which is difficult to achieve for CsI:Tl crystals even using high-quality finishing polishing, since it is an extremely plastic material. The main requirement for the output surface is its high transparency. To improve the energy resolution, the entrance and lateral surfaces are often matted [3]. In some cases, the output surface is also matted [4]. A common requirement for the surfaces intended for different functions is the time stability of their physicochemical states. The latter requirement contradicts the adopted matting techniques, since applying scratches

of a certain depth creates a diffusely reflecting relief. Such a matted surface is subject to strong polygonization, and the purposefully created relief spontaneously changes with time. For example, matting part of the lateral surface of a long detector is used to equalize the axial nonuniformity of the light output. According to the data from [5], the light-output uniformity falls outside the tolerable limits for 21% of finished articles because of unpredictable changes in the reflection conditions on the matted surface.

A specific feature of the CsI material is a low threshold for the start of dislocation glide [6]; therefore, even slight polishing causes a strain of the surface layer and generation of a large number of vacancies [7]. Vacancies create additional luminescent centers in CsI [8]; as a result, the scintillation efficiency near the surface is higher than in the crystal bulk [9] for 4–6 days after the polishing.

This paper presents a comparative analysis of different methods for treating the scintillator entrance surfaces illuminated by incident radiation. The objective of this study was to choose optimal techniques for manufacturing detectors with stable spectrometric characteristics (light output and energy resolution) during operation when detecting α particles.

EXPERIMENTAL

Samples of CsI:Tl crystals with dimensions of $\varnothing 25 \times 20$ mm (nos. 1 and 2) and $\varnothing 30 \times 5$ mm (nos. 3–7) taken for our measurements were cut from a common single crystal and had equal activator concentrations ($C_{\text{Tl}} = 4 \times 10^{18} \text{ cm}^{-3}$). This concentration is below the optimal one ($C_{\text{Tl}} = 1.1 \times 10^{19} \text{ cm}^{-3}$) for detecting α particles. The choice of C_{Tl} is conditioned by the results

from [7], where it was shown that the concentration of point defects in the near-surface layer after polishing reaches $C_D = 7 \times 10^{17} \text{ cm}^{-3}$. For this purpose, it is convenient to observe the effect of surface treatment on crystals with a relatively low activator concentration. The main conclusions of this study were also verified on crystals with the optimal activator concentration ($C_{\text{Tl}} = 1.1 \times 10^{19} \text{ cm}^{-3}$). Samples nos. 8–14 of CsI:Tl crystals with dimensions of $20 \times 15 \times 15 \text{ mm}$ were manufactured from a single crystal with activator concentration $C_{\text{Tl}} = 1.1 \times 10^{19} \text{ cm}^{-3}$.

The entrance surface for the incident radiation that was treated by different techniques was always oriented closely to the (110) plane. Samples nos. 1 and 2 were studied at two energies of α particles from the ^{239}Pu isotope— $E_1 = 5 \text{ MeV}$ and $E_2 = 2 \text{ MeV}$. Light output V and pulse-height resolution R were measured for two shaping times at the pulse-height analyzer— $\tau_1 = 1 \mu\text{s}$ and $\tau_2 = 2 \mu\text{s}$.

An R1307 (Hamamatsu) photomultiplier tube (PMT) served as a photoreceiver.

Samples nos. 3–7 were studied by detecting α particles with an energy of $E = 5 \text{ MeV}$.

Samples nos. 8–14 were studied by detecting both α particles with $E = 5 \text{ MeV}$ and γ quanta with $E = 662 \text{ keV}$ from the ^{137}Cs isotope.

All samples were mounted on the PMT without an optical contact. Special collimators ensured the required energy of α particles and their perpendicular incidence on the crystal surface. The instrumentation corresponded to the requirements of [10] imposed on the linearity and stability.

The entrance surfaces of CsI:Tl crystals (nos. 1 and 2) facing the radioactive source were first ground with a compound containing ethylene glycol and electrocorundum with $20\text{-}\mu\text{m}$ abrasive grains and then polished with ethylene glycol and aluminum oxide with $0.3\text{-}\mu\text{m}$ grains (methods 1 and 2). After completion of these measurements, these surfaces were subjected to the same grinding (but without polishing) and similar measurements were repeated. In addition to measuring the light output and the pulse-height resolution, relative specific light output $\eta = V/E$ and nonproportionality between the light output and the energy of α particles

$\Delta = \left(\frac{V_1}{E_1} - \frac{V_2}{E_2} \right) / \left(\frac{V}{\bar{E}} \right)_{\text{av}}$ were determined. This value characterizes the degree of deviation of the specific light output from a constant level.

The entrance surfaces of crystals nos. 3–7 were first subjected to a standard polishing procedure with water (method 3) and then to deep grinding–polishing. The surface was ground with a compound containing ethylene glycol and electrocorundum with abrasive-grain dimensions successively decreasing from 20 to $3 \mu\text{m}$. The coarse polishing was performed with a combination of ethylene glycol and aluminum oxide with

$0.3 \mu\text{m}$ grains. Subsequently, samples nos. 4–7 were treated using the following techniques:

(i) polishing with a compound containing methanol and a superdispersed zinc oxide powder (method 4, sample no. 4) and

(ii) polishing with a compound containing oligoorganosiloxan, tetraethoxysilan, ethanol, and a superdispersed silicon dioxide powder obtained by the sol–gel method (method 5, samples no. 5–7).

In the same experiment, we evaluated the stability of the samples' scintillation characteristics in time during their holding under normal conditions for 12 days.

All of the output surfaces of CsI:Tl crystals through which scintillation light leaves them with activator concentration $C_{\text{Tl}} = 1.1 \times 10^{19} \text{ cm}^{-3}$ and dimensions of $20 \times 15 \times 15 \text{ mm}$ were polished by the method 5, while the lateral surfaces and those facing the photoreceiver were treated according to the following procedures:

(i) samples nos. 8–10 were polished by method 5;

(ii) samples nos. 11–12 were treated by method 6—by a grinding compound containing a mixture of an oligoorganosiloxan-based liquid and tetraethoxysilan as a wetting fluid and M63 electrocorundum as a polishing powder;

(iii) sample no. 13 was treated by method 7—by a grinding compound containing ethylene glycol as a wetting fluid and an M63 electrocorundum as a polishing powder.

In the same experiment, we evaluated the stability of the samples' scintillation characteristics in time during their holding under normal conditions for three months.

RESULTS OF STUDIES

Table 1 lists the measurement results and the calculated characteristics of CsI:Tl crystals nos. 1 and 2.

Analysis of the experimental results presented in Table 1 shows that the specific light output increases after the surface is ground and a disturbed near-surface layer is formed (by $\sim 30\%$ at energy of α particles E_1 and by $\sim 60\%$ at energy E_2) compared to a polished surface.

Nonproportionality between the light output and the energy upon formation of a disturbed layer increases by $\sim 20\%$ and depends on signal-shaping time τ . At signal shaping time τ_2 , the nonproportionality magnitude is 3–5% larger. This indicates that the light output has increased mainly owing to the contribution of the slow scintillation component.

The pulse-height resolution does not virtually change with a change in τ . At the same time, its magnitude decreases by 1–3%, as a disturbed layer is created.

The light output of samples nos. 1 and 2 with the polished surfaces increases in the same proportion as for samples with ground surfaces, as τ increased by a factor of 1.25–1.27.