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ON THE SPECTROSCOPIC PROPERTIES OF HIGHLY DOPED CsI(Tl) SCINTILLATORS

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*Abstract***.** The results of a comparative study of the spectroscopic performances of the CsI(Tl) scintillator with Tl content between 0.1% and 0.2% are presented and discussed. Both energetic resolution and detection efficiency for alpha as well as gamma rays were experimentally determined for three 20 × 20 × 40 mm^3 CsI (Tl) crystals coupled with a PIN photodiode. The best energy resolution 3.8 % for ^{241}Am alpha rays and 2.8 % for ^{60}Co gamma rays were established in the case of a Tl content 0.15%, while the maximum detection efficiency was observed in the case of maximum 0.2 % Tl concentration

Key words: CsI(Tl), scintillator, highly doped, energy resolution, detection efficiency.

1. INTRODUCTION

CsI(Tl) is considered one of the brightest inorganic scintillators whose high wavelength of light emission of 550 nm makes it best coupled to red-enhanced photomultiplier tubes (PMT) or to photodiodes [1]. In the latter case, the use of a CsI(Tl) -photodiode pair significantly diminishes the size of detecting system (photodiodes are considerably smaller then PMT and need not high-voltage supply source) as well as the negative influence of magnetic fields. Moreover, CsI(Tl) is almost not hygroscopic and does not require an air-tight enclosure [2]. The only CsI drawback it related with its time resolution, estimated at 980 ns [3].

CsI scintillation properties such as energy resolution or detection efficiency are significantly influenced, in a complex manner, by the Tl content. At the same time, energy resolution depends on the CsI crystal quality, lower the defect content, higher the resolution [4–7].

Due to their compactness, the ensembles scintillators crystal + photodiode have found a great number of applications in nuclear spectroscopy, being used both for industrial [8–10] and scientific purposes [11–14].

Table 1

The main characteristics of Hamamatsu S3204-05 PIN photodiodes at 25° C

Parameter	Values
Active area	20 × 20 mm^3
Photo sensitivity	36% at λ = 540 nm
Spectral response domain	320 nm to 1100 nm with a maximum at 960 nm
Dark current	6.0 nA typical and 20 nA at V_R=10V
Terminal capacitance	130 pF at 1MHz and V_R=70V
Cut-off frequency	20 MHz at R_L=50Ω and V_R= 100V.

R_L = bias detector, V_R= bias voltage

Development at the National Institute for Physics and Nuclear Engineering "Horia Hulubei" of new experiments in nuclear physics involving the detection of a wide category of nuclear radiation, motivated us to perform a comparative study of the spectroscopic properties of CsI(Tl) scintillators with a Tl content varying between 0.1 and 0.2 % (1000 and respectively 2 000 ppm). Accordingly, we have experimentally determined both energy resolution and detection efficiency for a variety of radiaoctive sources including alpha, beta as well as gamma emitters.

The results thus obtained are presented and discussed further in this paper.

2. MATERIALS AND METHODS

1.1. DETECTORS

For our investigations we have used three CsI(Tl) scintillators having the same sizes, *i.e.* 20×20×40 mm^3, whose Tl doping content was of 0.1, 0.15 and respectively 0.2 %. All crystals, manufactured by Hangzhou Lambda Photonics Technology Co, Ltd. China, were sealed in a lightproof vacuum (10^{-3}–10^{-5} torr) chamber. To ensure a better collection of the scintillation photons, each crystals was optically connected to a 20 mm × 20 mm, large active area Hamamatsu S3204-05 PIN photodiodes [15] whose main parameters are illustrated in Table 1.

1.2. RADIOACTIVE SOURCES

In order to check both energy resolution and detection efficiency of the above mentioned detectors, we have used three different sources emitting alpha, beta and gamma rays whose characteristics are given in Table 2.

Table 2

Radioactive sources used in this study and their character*istics*.
All sources were small enough to be considered as point-like

Source	Radionuclides (Activity in Bq)
AMR 33 - R9207	^{239}Pu (2001), ^{241}Am (2024), ^{244}Cm (412)
SEA 4-2	^{241}Am (13000)
EG 3	^{60}Co 60 (37700)
SEB 7-3647	^{90}Sr+^{90}Y (2700)

1.2. ELECTRONICS

In all three cases, we have used the same electronic circuit that ensured both stabilized bias voltage and pulses amplification. Raw PIN signals were primarily processed by a charge sensitive preamplifier built in a SMT structure with discrete FET/BJT components at input and a fast operational amplifier at the output, similar to those described in ref. [16]. Further, signals amplified by a Canberra 2020 amplifier were recorded and analyzed by a built-in multichannel analyzer CAEN model N840 8k and a dedicated InterWinner 6.0 soft.

3. RESULTS AND DISCUSSION

In performing our investigations, we were interested to establish the optimum Tl concentration for all three types of scintillators as well as for all types of radiations for which this system was designed. Since energy resolution could be determined only by using monochromatic radiations, this characteristic were investigated only for alpha and gamma rays sources. The same thing was valuable for detection efficiency too.

For illustration, in Fig. 1 and 2 we have given the characteristics alpha lines of ^{241}Am (E_α = 5.486 MeV) as well of the triple alpha source AMR 33-R9207 (^{239}Pu: E_α = 5.245 MeV, ^{241}Am; ^{244}Cm: E_α = 5.805 MeV) as recorded by using each detector.

In the case of the SEA 4-2 ^{241}Am source, the alpha spectra were recorded for all three detectors in the same experimental conditions, *i.e.* source – scintillator distance of 0.1 cm and acquisition time of 2000 s, while for EG3, ^{60}Co, source we have maintained the same acquisition time but we have increased the source-detector distance to 20 cm.

In these conditions, the detection efficiency, g, was calculated by using the following equation [2]:

$$g = \frac{4\pi}{\Omega}\frac{R}{\Lambda}, \tag{1}$$

where: $\Omega = 4\arcsin\frac{l^2}{4(d^2 + l^2)}$ represents the solid angle subtended by detector; d represents source to detector distance; l is the detector side size; R is the counting ratio, Λ represents the source activity.

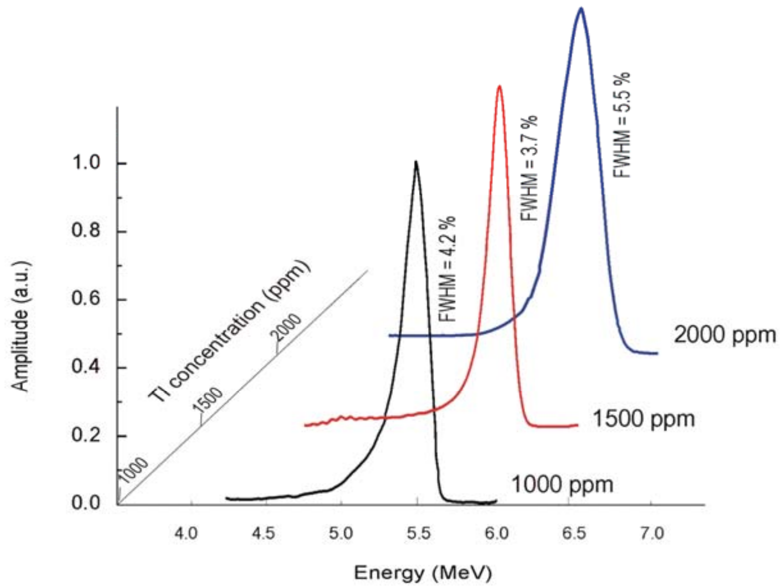

Fig. 1 – Energy spectrum of the 5.486 MeV line of ^{241}Am SEA 4-2 alpha ray source obtained by using CsI(Tl) crystals with different content of Tl. All spectra were obtained in the same experimental conditions, but for a better comparison all were normalized to maximum amplitude. It is worth mentioning in all cases the maximum amplitude corresponded to the same channel.

At a more detailed analysis of the data regarding detection efficiency (Table 3), it follows that the efficiency monotonously increases with the Tl concentration, in concordance with the role played by Tl ions in the process of excitons recombination followed by the emission of a light quanta. Accordingly, more Tl ions, higher recombination probability, which, at its turn determines an increased flux of scintillation photons for the same number of incident alpha or gamma rays, in concordance with the scintillation mechanism [4, 5]. Moreover, in all cases, the efficiency was systematically higher in the case of alpha ray than in the case of gamma ones, fact well explained by completely different mechanisms of interaction of charged and electromagnetic quanta.

Table 3

The experiential values of detection efficiency as a function of Tl concentrations

Tl concentration (ppm)	Efficiency (%)	
	^{241}Am (SEA 4-2)	^{60}Co (EG 3)
1000	91.3	41.29
1500	93.6	44.16
2000	94.5	46.30

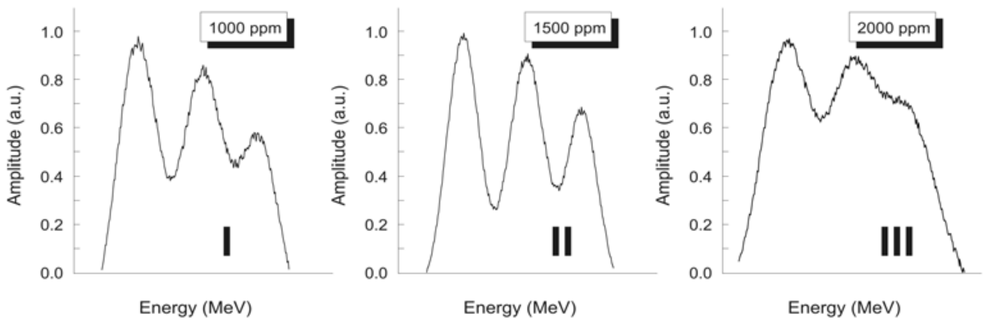

Fig. 2 – Energy spectra of the triple alpha source AMR 33 - R9207 (^{239}Pu, ^{241}Am; ^{244}Cm) recorded by means CsI(Tl) scintillators with different content of Tl. The 1500 ppm Tl detector shows the best energy resolution (spectrum II).

In the case of energy resolution, we have noticed a quite different dependence on the Tl concentration. Indeed, as it follows from the data given in Table 4, the energy resolution shows a minimum for both for alpha and gamma rays for a Tl concentrations of 1 500 ppm. In this case, initially the energy resolution defined as the ratio between spectrum line FWHM and its energy decreases with Tl concentrations up to 1 500 ppm and then increases reaching a maximum for 2 000 ppm.

These finding are better illustrated, by experimental spectra of an AMR 33 - R9207 composite source consisting of a mixture of three alpha emitters radionuclides with close energies: ^{239}Pu, ^{241}Am and ^{244}Cm (Fig. 2). It is evident that the best separation can be observed on the spectrum II, recorded by using a CsI(Tl) scintillator containing 1 500 ppm Tl.

The optimum of energy resolution noticed for 1 500 ppm Tl concentrations testifies for the best quality of these scintillators, since, as stated before, the energy resolution is significantly influenced not only by the Tl content but also by the multitude of defects induced during CsI crystals growth [4–7].

Similar results we have obtained in the case of SEB 7-3647 $^{90}Sr+^{90}Y$ beta rays source, where the highest measured detection efficiency corresponded also to 2 000 ppm Tl concentration.

Table 4

Numerical values of energy resolution as a function of Tl concentrations

Tl concentration (ppm)	Energy resolution (%)	
	^{241}Am (SEA 4-2)	^{60}Co (EG 3)
1000	4.2	6.7
1500	3.7	2.9
2000	5.5	5.8

In this way, all our results recommend the CsI(Tl) with 1500 ppm of Thallium as the optimum scintillator regarding both energy resolution and detection efficiency, point towards the necessity of a carefully selection of such kind of detectors before being used in nuclear spectroscopy.

4. CONCLUDING REMARKS

To select the best CsI(Tl) detectors for further spectroscopic applications, we have measured, in the same experimental conditions, both the energy resolution and the detection efficiency of three different CsI scintillators doped with Tl with concentrations between 1 000 and 2 000 ppm. By using ^{241}Am and ^{60}Co as standard sources, we have observed, for both types of radiation, that the detection efficiency increases with increasing the Tl content, reaching a maximum for 2 000 ppm, while the best values of energy resolution corresponds to 1500 ppm Tl concentration.

These results, confirmed by supplementary determinations for alpha ^{239}Pu, ^{241}Am and ^{244}Cm of as well as beta $^{90}Sr+^{90}Y$ ray source recommended 1 500 Tl ppm CsI(Tl) scintillators as the most appropriate one for further use in nuclear spectroscopy.

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