

PIXE characterization of CsI(Tl) scintillators used for particle detection in nuclear reactions

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Available online 6 March 2008

Abstract

Particle-Induced X-ray Emission has been used to measure Thallium concentration in several CsI(Tl) scintillators from different manufacturers, in order to check their nominal declared values and correlate their behaviour with actual Tl concentration. Indeed, both Tl doping level and its uniformity affect light emission of these detectors, which are largely employed in nuclear physics experiments.

In some of the examined crystals Tl concentration values from PIXE measurements came out to be quite different from those declared. This allowed us to explain apparent anomalies in the trend of their α/γ -induced light yield ratio versus Tl content. In some cases, the presence of unexpected contaminants was also pointed out.

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PACS: 82.80.Ej; 29.30.Aj; 29.30.Kv; 29.40.Mc

Keywords: Cesium iodide scintillators; Thallium doping; External PIXE

1. Introduction

CsI crystals doped with Thallium atoms are since decades employed in nuclear physics as charged particle detectors. They present high stopping power and good energy resolution (e.g. 3–4% FWHM for 5 MeV α particles) and are both easily machinable and rather cheap. In addition, particle identification is possible via pulse shape analysis [1], owing to the dependence of fluorescence emission on the kind of impinging particle. For these reasons, many modern arrays based on $\Delta E - E$ telescopes use CsI(Tl) crystals as residual energy detectors [2–4].

Recently, an international collaboration (FAZIA) [5] started an intense R&D program aimed at optimizing the

performance of the commonly used nuclear detectors, in view of the construction of a new large array for heavy ion physics in the next generation facilities for exotic beams. Concerning CsI(Tl), FAZIA plans in particular to check whether energy resolution, strictly connected with the scintillation yield, improves for heavy ions at high Tl concentrations. As in all inorganic crystals, yield and spectrum of fluorescence light emitted by CsI(Tl) detectors when hit by radiation crucially depend on the doping amount and on its homogeneity; basing mainly on scintillation studies for γ and α particles, it was observed that light response tends to saturate at Tl concentration values greater than 1000 ppm [6]. Therefore, manufacturers generally produce crystals with Tl doping around 1000 ppm. However, it is not clear whether the light yield still increases for heavy fragments when CsI is over-doped with Tl.

In order to investigate this issue, the FAZIA collaboration got a number of crystals with different nominal Tl content, also purchased by different brands. As a first step in the planned tests, it was decided to directly verify by

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PIXE analysis the declared Tl content, as it is obviously important to know its actual value when comparing results from different crystals. At the same time, PIXE appeared to be suitable also to detect possible spurious dopants in the CsI matrices, the presence of which, even in trace amounts, can affect light response. We present and discuss here PIXE results obtained in the framework of these investigations for some CsI(Tl) crystals, with parallelepiped shape and dimensions $20 \times 20 \times 40 \text{ mm}^3$.

2. CsI performance and Tl concentration

An easy and common way to characterize the response of inorganic scintillators to heavy charged radiation is to measure the ratio between the pulse heights for α particles and γ of the same energy [7], i.e. the so called α/γ ratio. Usually, this is done in the laboratory using calibration sources. As mentioned above, the α/γ ratio has been reported to increase as Tl concentration increases up to values of about 1000 ppm, then it tends to saturate [6,7]. Fig. 1 shows the results we obtained for the α/γ ratio of a group of detectors we had at disposal for the FAZIA tests, plotted versus the nominal Tl concentration declared by the manufacturers. Measured values (filled circles in the figure) are compared with data from previous works by other groups (line-connected stars); it is apparent that our experimental points do not follow the trend reported in the literature and appear widely scattered. This might be explained by a wrong abscissa for the data referring to some crystals; it was thus decided to verify the actual Tl concentration by direct measurements.

3. PIXE results

3.1. Tl concentration

Tl doping concentration was measured by PIXE, exploiting the standard two-detector set-up [8] installed at one of the external beam-lines of the 3 MV Tandatron accelerator at LABEC, Florence. The bare CsI crystals were easily placed in front of the beam exit window. Irradiation was performed with a 2.9 MeV proton beam (about $85 \mu\text{m}$ range in CsI) at a current of about 250 pA; each run lasted from 200 to 1000 s (depending on the Tl amount dispersed in the matrix). One of the two X-ray detectors, a Si(Li), collected X-rays over a wide solid angle (about 100 msr), and it was provided with a proper absorber ($430 \mu\text{m}$ Mylar + $36 \mu\text{m}$ Al) to drastically reduce the otherwise too large count-rate of I and Cs L-X-rays in the region 3.9–4.9 keV, while still keeping a good transmission for the Tl L-X-rays at 10.2–12.2 keV. A second detector with a high transmission of low-energy X-rays (but covering a much smaller solid angle) was used to check possible contaminations also from low-Z elements; the major information was however derived from the data of the larger solid angle detector (therefore only spectra from this detector are reported in the following).

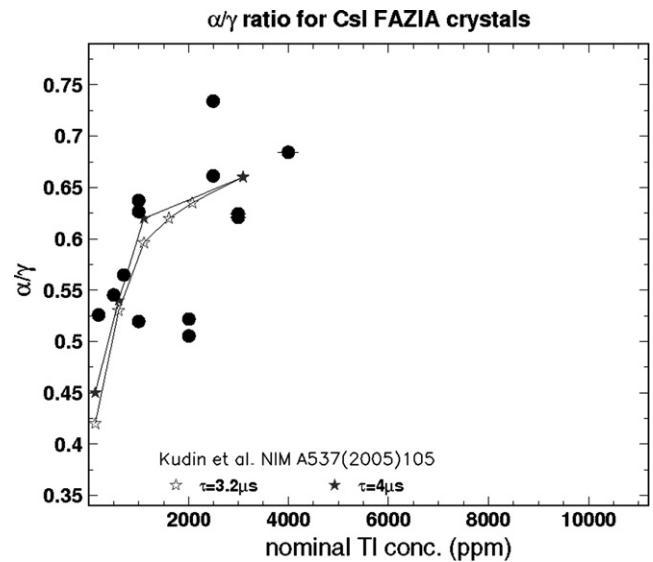


Fig. 1. α/γ ratio (normalized to the ^{241}Am α -energy) as a function of Tl content in CsI scintillators: the measured values (filled circles, obtained with shaping constant $4 \mu\text{s}$) for the crystals under study in this work are reported versus their nominal Tl concentration and compared with data from literature (line-connected stars).

The beam spot was only about 0.3 mm^2 , thus, for each crystal we performed at least an average measurement (AM), scanning a central 1 cm^2 area on one of the square faces of the detector. For some crystals we irradiated the two square faces (two AMs) while for a few scintillators we also tested the lateral sides and/or performed spot measurements (SM), e.g. close to the corners, in order to sample local Tl concentration and check for its homogeneity. The spectra were analysed using GUPIXWIN, the most recent version of the GUPIX software [9,10].

Results showed strong discrepancies between nominal and actual Tl content (quantified from AMs); in addition, for some crystals, relevant differences between punctual and average values were also pointed out. An example is shown in Fig. 2, where the PIXE spectra for two crystals with different Tl concentration are reported, normalized to the area of the K_{α} line of iodine. The insert of Fig. 2 clearly shows even visually that the different yields of the Tl characteristic L-lines do not reflect at all the ratio of the nominal Tl concentration values, namely 3000 and 2000 ppm. From GUPIX analysis, indeed, the measured Tl concentration for the latter crystal comes out to be largely different ($580 \pm 40 \text{ ppm}$, AM value) from the nominal value.

In Fig. 3 an example attesting also the non-homogeneity of Tl doping over the examined surface of one of the crystals is reported. PIXE spectra (normalized to the iodine K_{α} peak area) obtained from the central 1 cm^2 area of the scanned face and from a lateral spot are compared: a large difference in Tl content can be observed in the two cases (AM $\sim 6400 \text{ ppm}$, SM $\sim 9300 \text{ ppm}$).

The whole of the PIXE results for Tl concentrations of all the analysed crystals allowed us to reposition the α/γ

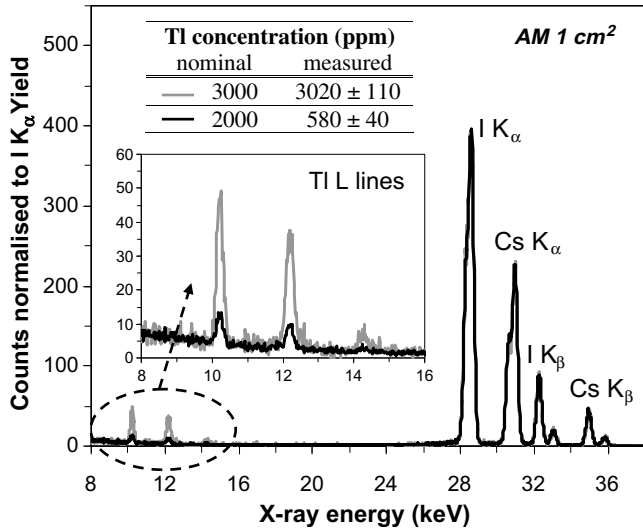


Fig. 2. PIXE spectra (normalized to iodine K_{α} peak area) for two crystals with different TI concentration. For one of them the actual TI content is largely different from the nominal value. The spectra refer to average measurements (AMs), performed by scanning a 1 cm^2 area.

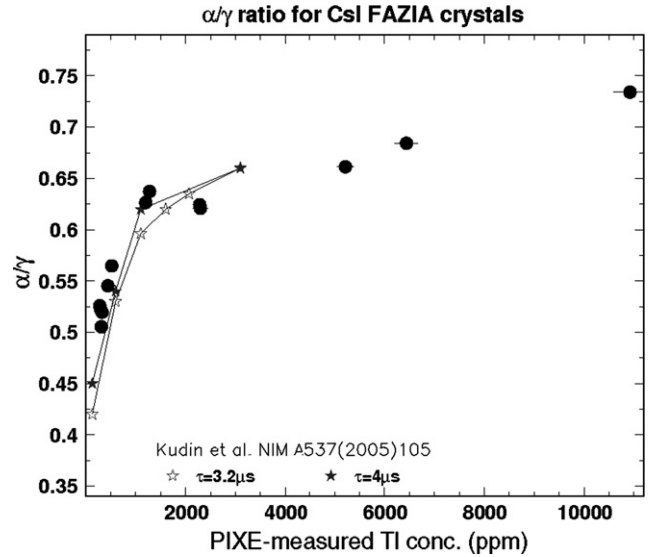


Fig. 4. The measured α/γ ratios (filled circles) of Fig. 1, repositioned using as abscissa the PIXE-measured TI concentration. Line-connected stars refer to data from literature.

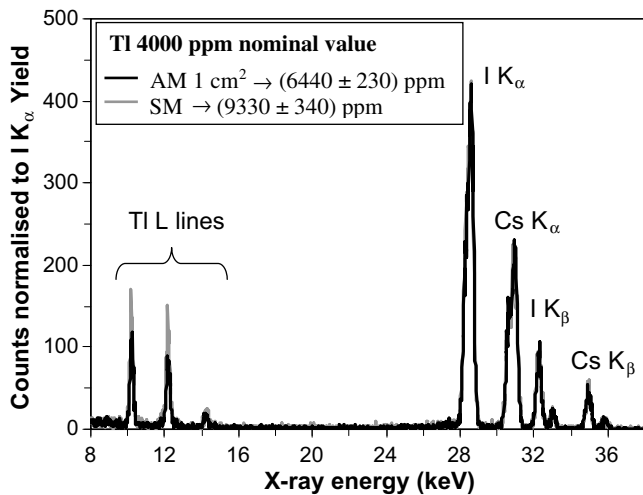


Fig. 3. Comparison between PIXE spectra (normalized to iodine K_{α} peak area) obtained from the central 1 cm^2 area of the scanned face (AM) and from a lateral spot (SM) for one of the CsI(Tl) crystals analysed.

ratios of Fig. 1, using as abscissa the PIXE-measured (AM values) rather than the nominal concentration. The effect of this operation is presented in Fig. 4: a good agreement with the expected smooth trend is now achieved for all our crystals.

3.2. Spurious element contamination

The presence of unwanted impurities in the crystal can modify light emission of inorganic scintillators, thus affecting their performance in terms of energy resolution. Among the examined crystals, contamination of unexpected trace elements was indeed found in those, which had shown bad energy resolution for γ and α particles.

An example is shown in Fig. 5, which reports the PIXE spectra relative to two crystals with 1000 ppm nominal TI content, and identical as to their optical appearance. However, one of them (A) had shown good energy resolution while the other (B) a very bad one (e.g. the three lines of the α particles of the calibration source were unresolved). While the PIXE-measured TI amount results to be reasonably compatible with the declared value for both, in sample B the presence of bromine can be observed, at a moderate level of concentration (200 ppm). Other crystals were found to be contaminated with relevant amounts of iron (e.g. two crystals from the same supplier came out to have about 700 ppm Fe concentration).

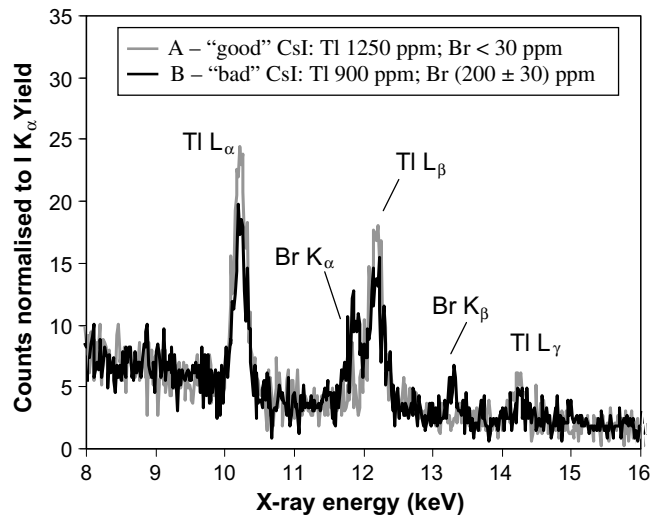


Fig. 5. PIXE spectra for two crystals (TI 1000 ppm nominal): scintillator (A) had shown good energy resolution, while (B) a very bad one. In sample (B) the presence of bromine can be observed as a contaminant.

These preliminary measurements suggest a possible correlation between contamination and energy resolution worsening, which however should be more extensively investigated for a definite assessment.

4. Conclusions

PIXE demonstrated to be a useful tool to check the concentration of doping species dispersed in inorganic crystals. Although it cannot explore the whole bulk of the detectors, PIXE can readily be used for a quick check of the first tens of microns from crystal faces. The results on Tl concentration in CsI crystals that we obtained by PIXE allowed us to take back the α/γ ratio versus Tl concentration to the trend expected from literature. We plan therefore to extend these investigations in the future to new crystals of the FAZIA collaboration for which the Tl content issue is crucial.

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