HNPS Advances in Nuclear Physics

Vol 1 (2025)

ANP2024 Special Volume

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doi: 10.12681/hnpsanp.8134

BEGAM, a new apparatus for the identification of β emitter contaminants in radiopharmaceuticals

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Abstract The BEGAM project aims at developing a portable detector capable of performing γ/β coincidence and anti-coincidence measurements to estimate the presence of long-lived β-contaminants in radiopharmaceutical solutions. A preliminary prototype of the apparatus is under construction. It is composed of four sectors of Caesium Iodide (CsI) scintillators surrounding a cylindrical plastic scintillator. The scintillation readout is performed via silicon photomultiplier (SiPM) and after some preamplification the signals are sent to a commercial digitizer. Geant4 simulations of the detector's response have been developed and validated using gamma and electron sources. Preliminary γ/β coincidence measurements from a ^{207}Bi source have been performed. Near-future plans include testing the BEGAM detector to estimate the activity of ^{99}Mo contaminants in ^{99m}Tc solutions.

Keywords Radiopharmaceuticals, scintillator detectors, gamma-ray spectroscopy, β-tagging

INTRODUCTION

Radiopharmaceuticals are specialized drugs used in nuclear medicine, composed of a radioactive isotope bonded to a carrier molecule with suitable biochemical properties [1]. These drugs use the radiation emitted by the isotope for medical applications. Depending on the isotope's decay mode, radiopharmaceuticals are used for either diagnostic or therapeutic purposes. For diagnostics, radiopharmaceuticals typically utilize isotopes that emit suitable γ radiation or undergo positron decay (β+), such as ^{99m}Tc [2] or ^{18}F [3]. The radiation emitted allows for imaging techniques like Positron Emission Tomography (PET) and Single-Photon Emission Computed Tomography (SPECT), enabling precise visualization of internal structures in the human body. In contrast, therapeutic radiopharmaceuticals use isotopes that decay via β- or alpha (α) emissions to target and destroy specific cells, like cancerous tissues [4].

Radioisotopes for radiopharmaceuticals can be produced through various methods, including cyclotrons [5], nuclear reactors, and nuclide generators [6]. While these methods are all effective, they inevitably introduce trace contaminants into the final solution. Therefore, ensuring the purity of radiopharmaceuticals is crucial to minimize potential risks to patients. Taking ^{99m}Tc as an example, its production from ^{99}Mo in radionuclide generators requires a careful estimation of residual ^{99}Mo contamination. For this type of measurement, a cylindrical ionization chamber is used, also known as dose calibrator. This allows for estimation of the contamination amount of the eluate, but only in a semi-quantitative way. The BEGAM project focuses on techniques for identifying contaminants in radiopharmaceuticals employing scintillator detectors. The approach involves coincidence and anti-coincidence measurements of γ and β radiations to accurately estimate the activity of long-lived β-emitting contaminants.

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EXPERIMENTAL DETAILS

The goal of the BEGAM project is to develop a portable detector capable of performing coincidence and/or anti-coincidence measurements on radioactive solutions, specifically radiopharmaceuticals, to accurately estimate the presence of long-lived β-contaminants. Here the performances of the first prototype of the BEGAM detector are described. In this design, four cylindrical slices of cesium iodide (CsI) surround a plastic scintillator. Each CsI sector measures 50 mm in height with a thickness of 133 mm (Fig. 1a). Two different plastic scintillators are used, both with a height of 50 mm and an external diameter of 33 mm; however, one has a thickness of 4 mm (Fig. 1c), while the other is 13 mm thick (Fig. 1b).

The thicker plastic scintillator ensures the complete absorption of electrons with energies up to 2 MeV. However, in the choice of the thickness also the sensitivity of the plastic scintillator to γ-rays should be taken into account. Efficiency values for γ-rays at various energies for the two thicknesses have been obtained through Geant4 simulation. Results show that the percentage of γ-rays interacting with the plastic scintillator via Compton scattering or the photoelectric effect is around 3% for the 4 mm thickness and 9% for the 13 mm case. This means that a considerable number of γ -rays may be absorbed or may reach the CsI detector only after undergoing a Compton scattering in the plastic scintillator, thus originating a huge background in the coincidence spectra.

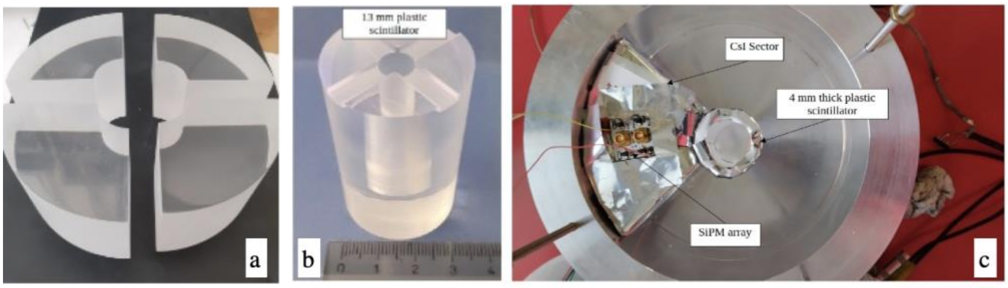

Figure 1: *Panel a): the four CsI sectors of the BEGAM prototype. Panel b): the 13 mm thick plastic scintillator c) The setup used in the test measurements.*

The light produced by the scintillators is collected using silicon photon multipliers (SiPMs). Each CsI scintillator is equipped with an array of four SiPMs, positioned approximately at the center of the top face (Fig. 1c). The plastic scintillator instead is read by a single SiPM placed on its bottom face.

RESULTS AND DISCUSSION

To test the possibility to acquire coincidence spectra with BEGAM we performed a γ-electron coincidence measurement using a radioactive source of ^{207}Bi. The ^{207}Bi source emits two intense γ -lines at an energy of 570 keV and 1064 keV, respectively. These γ-rays are emitted sequentially within a few picoseconds. Additionally, there is a significant probability that these transitions occur via the emission of conversion electrons with energies of approximately 975 keV and 481 keV, respectively. The 1 MeV conversion electrons are emitted in cascade with the 569 keV γ rays.

So far, the test has been made with a single sector of CsI scintillator was used alongside a half-cylinder plastic scintillator with a thickness of 4 mm. The half-cylinder is used to permit measurements with the solid ^{207}Bi source instead of the radiopharmaceutical eluate.

Fig. 2 presents the spectra obtained from the CsI scintillator (left panel) and the plastic scintillator (right panel) placed in front of it. Since according to our simulations, the efficiency of the plastic scintillator is rather low for 1 MeV γ-rays (about 3%), the peak around channel 1000 corresponds to the conversion electrons from the 1064 keV transitions.

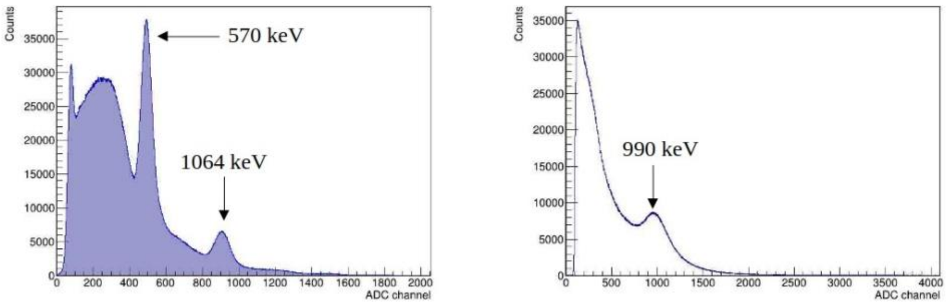

Figure 2: *Single spectra obtained from a radioactive source of* ^{207}*Bi obtained with the CsI scintillator (left panel) and with the plastic scintillator (right panel) The two peaks in the left panel corresponds to the γ-rays of the 570 keV and 1064 keV, respectively.*

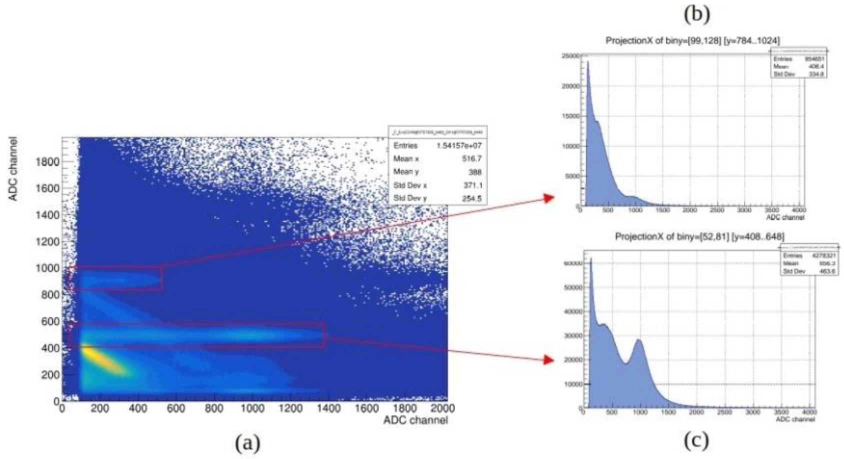

Figure 3: *2D coincidence histogram displaying the energy recorded by the plastic scintillator on the x-axis and the energy detected by the CsI scintillator on the y-axis (a). On the right, two x-projections are shown: (b) a projection taken outside the coincidence region and (c) a projection within the coincidence region.*

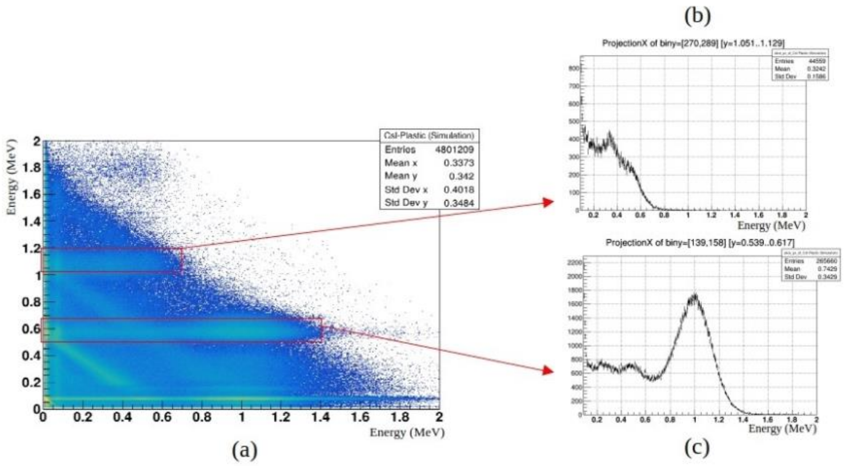

Figure 4: *2D coincidence histogram displaying the energy recorded by the plastic scintillator on the x-axis and the energy detected by the CsI scintillator on the y-axis according to the Geant4 simulation(a). On the right, two x-projections are shown: (b) a projection taken outside the coincidence region and (c) a projection within the coincidence region.*

We then acquired the signals from the two detectors imposing a coincidence condition. The resulting histograms are shown in Fig. 3. Fig. 3a displays a 2D histogram: the x-axis reports the energy (in channels) from the plastic scintillator and the y-axis the energy (in channels) from the CsI scintillator. Figs. 3b and 3c are projections onto the x-axis, corresponding to the two distinct regions of the 2D histogram highlighted by red rectangles. The projection corresponding to the region around channel 500 in the CsI detector (i.e. on the 570 keV transition) clearly shows a peak in the region where the 975 keV conversion electron line is expected in the spectrum of the plastic scintillator. These results confirm, so far qualitatively, the possibility to perform coincidence measurements with the BEGAM apparatus.

The results have been tested against a Geant4 simulation (Fig. 4). The coincidence histogram obtained from the simulation is close to the real case and the projection of the coincidence area shows that the plastic scintillator detects electrons around the same energy region as the real case.

CONCLUSIONS

At present, we are utilizing an intermediate prototype of the BEGAM detector, which consists of CsI scintillators for γ spectroscopy and a plastic scintillator for β spectroscopy. This prototype has undergone extensive testing through Geant4 simulations and experimental validations. The simulations revealed that a high thickness of the plastic scintillator could pose challenges in performing β-γ coincidence measurements, as a non-negligible fraction of low-energy γ -rays can interact with the plastic scintillator. This interaction may negatively impact the estimation of contaminants in widely used radiopharmaceutical solutions which emit low energy γ -rays, such as ^{99m}Tc, whose most prominent γ emission occurs at 140.5 keV.

Experimental measurements conducted using a ^{207}Bi radioactive source have validated the detector's capability for coincidence spectroscopy. The coincidence analysis successfully identified the expected signals from conversion electrons and their sequentially emitted γ-ray emissions, thereby demonstrating the feasibility of the system for isolating and quantifying contaminants.

Additionally, light collection studies for the CsI scintillators, both simulated and experimental, were carried out by varying the number of active SiPMs. The close agreement between simulated and experimental results confirms the reliability of the Geant4 simulations. These simulations will serve as an important tool for optimizing the design of future BEGAM detector prototypes.

In the near future, the BEGAM detector will be used to quantify the activity of ^{99}Mo contamination in ^{99m}Tc solutions. The samples will be provided by the "Azienda Universitaria Ospedaliera Careggi" (AOUC), which is collaborating closely with us on the development of the detector.

References

[1] S. Zhang, et al., Signal Transduct. Target. Ther. 10, 1 (2025); doi: 10.1038/s41392-024-02041-6
[2] J. Steigman, et al., Sem. Nucl. Med. 4, 269 (1974); doi: 10.1016/S0001-2998(74)80014-0
[3] D. Bars, J. Fluor. Chem. 127, 1488 (2006); doi: 10.1016/j.jfluchem.2006.09.015
[4] E. Akgun, Fabad Journal of Pharmaceutical Sciences 46, 93 (2021)
[5] D. Soloviev et al., Cyclotron Produced Radionuclides: Guidance on Facility Design and Production of [18F]Fluorodeoxyglucose (FDG), IAEA (2012)
[6] Non-HEU Production Technologies for Molybdenum-99 and Technetium-99m, IAEA Nuclear Energy Series (2013)