GAMMA-RAY SPECTROMETRIC CHARACTERIZATION OF WASTE ACTIVATED TARGET COMPONENTS IN A PET CYCLOTRON *

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Abstract

Irradiation of a target in a PET cyclotron creates high activity induced species in its components. In this work, a nuclide identification and activity evaluation of some activated target parts by high resolution gamma-ray spectrometry with HPGe detectors has been performed. Because of the high activity of some target components, a suitable collimation lead-walled system has been realized. Such system allows us to realize a gamma-ray spectrometric measurement of a small element of the target part at a remarkable distance from the detector, to put in comparison with the one of a calibrated point source of ^{152}Eu. Measurements on some target body components used inside an IBA Cyclone 18/9 cyclotron, allow the identification and activity evaluation for the most important radionuclides produced by activation of the materials.

INTRODUCTION

Positron Emission Tomography (PET) has become an imaging technique widely used in the world as both a morphological and a functional diagnostic tool. For these aims, a radioactive substance needs to be administered to the patients, generally a short-lived positron emitter produced with a cyclotron. One of the most important problems in using a PET cyclotron concerns the high activity induced in the components, in particular in the target body. The radionuclide inventory in the various parts is mainly determined by interactions of proton beam or secondary neutrons produced in the target. The problem is important and involves radiation protection issues when operators are engaged in maintenance or replacement of a target or its components. In fact, most of the target body elements are frequently replaced and generally stored in a Pb-shielded container in order to wait for their radioactive decay. Before the storage, it is required to report the activity values of the most important radionuclides present in the component. In fact, the removal of radioactive waste target body components is performed by an authorized company on condition that the main radionuclide species and their activities are known. Studies on residual radioactivity induced in a target after an irradiation cycle were already presented in [1,2,3], the last aimed also to evaluate maintenance personnel radiation exposure. In this work we perform a nuclide identification and activity evaluation of some activated target body parts by high resolution gamma-ray spectrometry with HPGe detectors. Most of the components, after a suitable decay period, can be measured at high distance from the HPGe detector. To measure target parts whose activity is very high, i.e. fresh-irradiated target windows, a suitable collimation lead-walled system has been realized. Such a system avoids problems related to high measurement dead-time and reduces maintenance service operators exposure, too. Measurements on some stripper forks, carbon foils, Havar® foils and titanium vacuum window of a target used inside an IBA Cyclone 18/9 cyclotron, allow the identification and activity evaluation for the most important radionuclides produced by activation of the materials (^{48}V, ^{46}Sc, ^{51}Cr, ^{52}Mn, ^{54}Mn, ^{56}Mn, ^{56}Co, ^{57}Co, ^{58}Co and others).

MATERIALS AND METHODS

The IBA Cyclone 18/9 cyclotron used in the "S. Gaetano" Nuclear Medicine Center in Bagheria (a town near Palermo) was installed in May 2002 and started its ^{18}F-fludeoxyglucose ([^{18}F]FDG) production in January 2003. The Cyclone 18/9 is capable of accelerating negative hydrogen ion (H-) or deuteron (D-) to energies of 18 MeV and 9 MeV respectively. The cyclotron has been used for the production of [^{18}F]FDG via ^{18}O(p,n)^{18}F reaction into a niobium (or silver) target cell filled with about 0.5 ml (small target) or about 2.2 ml (large target) of [^{18}O]enriched water. The target was typically irradiated for about 60 min with 18 MeV protons and an average current of 32 μA. The main target components are generally replaced, except for failures or anomalies, after about 140 h (integrated irradiation time). This figure has been achieved after the use of Niobium in place of Silver as body target material.

The replaced parts are stored in a 5-cm thick lead-walled container, placed within a controlled area because the emerging dose level is about 100 times higher than the ambient background dose rate. Havar® foils, stripper double forks, carbon foils and titanium vacuum window of target irradiated during a 2-months production cycle (integrated target current: 1440 μAh) were analized by high resolution gamma-ray spectrometry. In order to identify short-lived induced nuclides, a new stripper device (named St1) was mounted and extracted after a few hours after the end of irradiation.

All the spectrometric measurements were performed using an HPGe (High-Purity Germanium) detector manufactured by ORTEC™ model GEM18180 (relative efficiency 18%, energy resolution 1.8 keV at 1332 keV) connected to an ORTEC mod. 672 Amplifier and an ORTEC 919E MultiChannel Buffer into an Ethernet

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environment. Data analysis was realized by ORTEC Gamma-Vision® Software (version 6.06). Energy and efficiency calibrations were performed by measuring standard sources of ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{113}Sn, ^{137}Cs, ^{54}Mn, ^{65}Zn (single line sources) and ^{152}Eu. The first sources, furnished by CEA-LMRI, were employed for close geometries (source-detector distance less than 10 cm), whereas ^{152}Eu source (furnished by Amersham) was used in far geometries (25 cm and more) and in collimated geometry. Energy and gamma-ray emission probability values were taken from [4].

Measurements of high specific activity fresh-irradiated Havar® foils (50 μm thick, 30 mm in diameter) or titanium window (12.5 μm thick, 24 mm dia.) give origin to a high measurement dead-time even when very large distances from the detector were used. Low dead-time and acceptable count-rate must be achieved in order to improve the spectrometric system performances. For this purpose, a suitable collimation lead-walled device has been realized in order to reduce the measurement dead-time and also as a protective shielding for the operators. In fact, dose rate value measured by a Berthold TOL/F ionization chamber, at a distance of 10 cm from an Havar® foil, was about 6 mSv h^{-1} after a decay period of about 25 days from the last irradiation (41 μSv h^{-1} at 1 m). For stripper double forks, after a decay of only 1 day, dose rate value was considerably lower (about 60 μSv h^{-1}). Such collimation system allows us to measure a small element of the target component, regarded as a point source, at a remarkable distance from the detector and to compare the results with those of a calibrated point source of ^{152}Eu. Such method represents a valid option to the complex dissolution in aqua regia procedure presented in [2]. The total activity can be obtained by multiplying the value related to the measured source fraction area (about 1cm^2) by the real source area, assuming that activity is uniformely distributed. The last hypothesis can be verified by measuring different area parts of the source and comparing the count-rate results. Otherwise, measurements on different source area fractions are required. Figure 1 reports a schematic section of the collimation device.

RESULTS AND DISCUSSION

Table 1 reports the main identified radionuclides besides the evaluation of the activities at the time of the last irradiation. We note that the total activity of waste stored in the box container is nearly entirely related to Havar foils, whereas other pieces were less important. Moreover, the largest contribution to activity is related to radionuclides with half-life of about 70 days. This means that after approximately 2 years (about 10 times the half-life), the total activity will be reduced to less than 0.2 MBq (reduction factor: 1000) and the worker can operate for waste disposal with a relative safety. The radionuclide activity values are close to those reported in [1], which confirms the suitability of the method, with the only exception of ^{51}Cr and ^{57}Co lower activity values. Figure 2 shows the measured gamma-ray spectrum detected on a Havar® foil extracted after a 2-months cycle of irradiation and stored for about 25 days. Although the collimation lead-device has been used, we were not able to reduce the measurement dead-time to less than 5%. For the other components, only the first measurements on the titanium target vacuum window were realized with the collimation device, whereas for the other ones a large source-detector distance has been satisfactory employed. Figure 3 shows a portion of gamma-ray spectrum detected on titanium vacuum window after a sufficient decay time in order to avoid the use of collimation device (122 days).

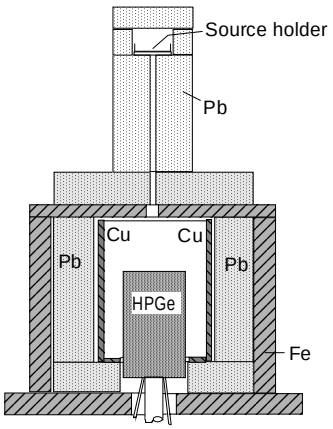

Figure 1: Layout of collimation device (Not to scale).

Table 1- Activity values in various target components corrected for decay to the time of the last irradiation.

Isotope	Half-life (days)	Activity (MBq)		
		Havar foil	Titanium window	Stripper fork
^{46}Sc	83.83	<DL	0.74	<DL
^{48}V	15.976	<DL	30.2	<DL
^{51}Cr	27.704	0.45	<DL	0.009
^{52}Mn	5.591	29	<DL	<DL
^{54}Mn	312.20	0.8	<DL	0.0014
^{56}Co	77.7	24	0.031	0.17
^{57}Co	271.77	0.4	0.00064	0.003
^{58}Co	70.916	65	<DL	0.0012

(DL=Detection limit for proper measurement geometry)

Other short-lived radionuclides can be recognized in the spectra detected on the stripper fork (St1) extracted after about 1 day after the irradiation (minimum time to permit operator access into the cyclotron vault room). We point out attention on the identification of ^{55}Co and ^{57}Ni, relatively short-lived nuclides produced by proton interactions. The other radionuclides reported in [1], as ^{60}Co or ^{59}Fe, cannot be recognized because of their low activity or short decay period. Measurements performed with various decay time permit to verify the values of all half-times and to identify long-lived nuclides, as for example ^{65}Zn. Nevertheless, they contribute little to the activity evaluation. Figure 4 reports a comparison of two gamma-ray spectra detected on St1 with a decay time of 1 and 44 days.

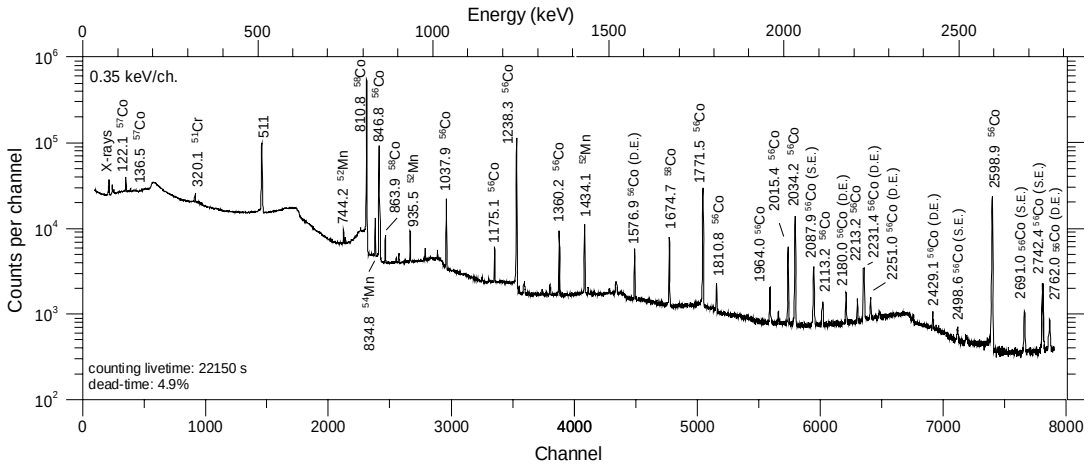

Figure 2 : Gamma-ray spectrum detected on a Havar® foil with lead-walled collimation system. (Decay time: 25 days; S.E.= single-escape peak; D.E.= double-escape peak).

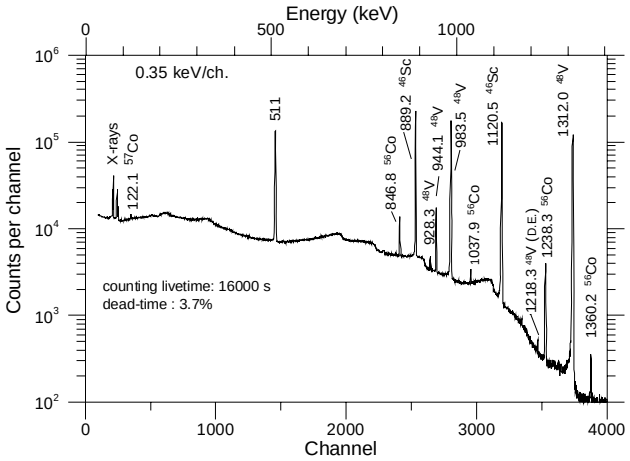

Figure 3: Gamma-ray spectrum detected on titanium window without lead collimation system. (Source-detector distance: 25 cm; decay time: 122 days).

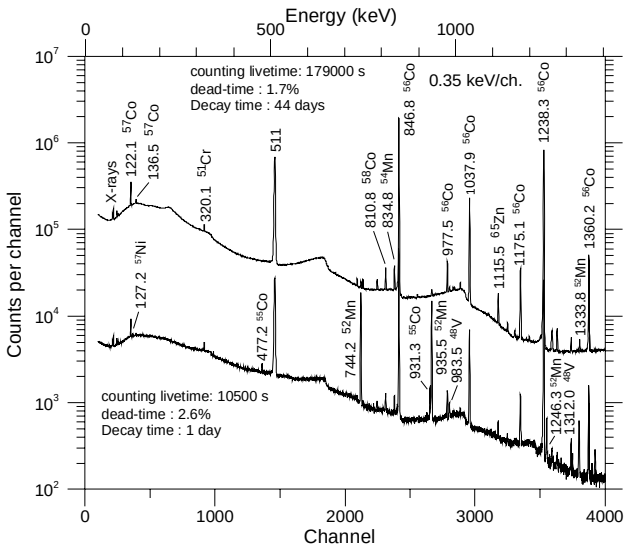

Figure 4: Gamma-ray spectra detected on stripper double fork (St1) extracted after a few hours from a irradiation. Source-detector distance: 25 cm.

CONCLUSIONS

Gamma-ray spectrometric measurement of activated target components is an useful technique to evaluate the nuclide activities in order to fulfil radiation protection requirements. Measurements can be easily performed in a PET cyclotron plant by adjusting an HPGe spectrometric system, with a limited initial outlay. Such a system allows us also to verify residual activity levels of cyclotron and shielding components. A collimation lead-walled device can be a suitable solution to measure very high induced activity target components and also to reduce maintenance service operators exposure.

Since the most important contribution to waste activity is related to radionuclides with relatively short decay time, we can confirm that after a decay period of about 2 years the waste components represent a rather low radiation hazard. In order to fulfil the last requirement, at least two Pb-walled containers seem to be required. In the first, with a shielding lead wall at least 10 cm thick, the most recent replaced parts must be stored in order to reduce the total activity level whereas in the second the oldest components are kept.

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