Analysis of radioisotopic metal impurities generated in [18O]H₂O during 18F production with cyclotron

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Abstract - In this work we present a characterization of the radionuclidic impurities originated by proton irradiation of enriched water [18O]H₂O in a medical cyclotron through Monte Carlo simulations and experimental measurements. A set of standard samples of enriched water loaded in the cyclotron target cell have been irradiated at 30 μA proton current for one hour each and, after an appropriate cooling time, measured by HPGe gamma spectrometry. In this way it was possible to study the direct release of radionuclidic impurities from target components as well as the release in the form of target ageing. Previously to experimental measurements, Monte Carlo calculations with the Pits Code have been carried out to estimate the radionuclides generated within the reactor components. These calculations were performed with the Monte Carlo Code MCNP (Ver. 5C), developed at the Los Alamos National Laboratory. The interaction of protons with the target material is described by the new 2C fissile target model. The relative efficiency of 25–30% and a resolution of 1.95 keV FWHM at 511 MeV (with a peak to Compton ratio of 55/1). MAESTRO® multi-channel Analyzer (MCA) and the detector to the HPGe-based HPGe detector. Gamma Vision® software has been used for peaks identification and evaluation. High resolution gamma spectrometry results: the comparison simulation and experimental results (Figure 8) shows that the amount of nuclides produced in Havr® window are several orders of magnitude lower than the ones measured in the 30 μA irradiated water, indicating that a very small portion of material is released from the Havr® foil to the irradiated enriched-water volume during the bombardment process following erosion/corrosion effects. This process tends in general to reduce with time, irradiation after irradiation. This effect can be explained as the effect of the removal of surface metallic residues at each irradiation mixed with the phenomena of trap-ping the produced nuclides by the surfaces of the target cell. Higher initial release of nuclides can be avoided by carefully cleaning all the spare parts involved in target cell maintenance and keeping all the irradiation condition as stable as possible could reduce the probability to introduce further effects on cell target. The study presented here can be further extended and applied to the assessment of radionuclidic purity in case of new target materials directly in contact with the irradiated volume.

Keywords: Analysis of radionuclidic impurities, [18O]H₂O, gamma spectrometry, MCNP, Monte Carlo simulation.

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Introduction

- Cyclotron-based radionuclide production and chemical synthesis of the marked molecule constitute the two main processes of the production of the most common radionuclides for Positron Emission Tomography (PET) (Aldaudin, 2012) such as [18F]-2-fluoro-2-deoxy-D-glucose ([18F]FDG). In the production process, (p,n) nuclear reactions induced by 18 MeV proton beam into a target which contains the [18O]-enriched water sample give rise to the production of [18F] (IAEA, 2009).

- Interactions of protons with the materials of the target cell and material activation due to the neutron secondary radiation field produced by the (p,n) reaction (IAEA, 2011), causes the formation and release of radionuclide pollutants during the irradiation process (Bowden et al., 2009; Gillies et al., 2006).

Fluorine-18 production with cyclotron

- The cyclotron installed at the Applied Nuclear Energy Laboratory (LENA) of the University of Pavia, is an IBA cyclotron (model Cyclone® 18/9) set up for 18 MeV proton bombardment of a highly PO-enriched H₂O target [18O=98.0 Atom %, provided by Huyi Isotopes Co.] with a nominal 30 μA beam current.

Formation of radionuclides inside the target cell

- During proton bombardment the target cell (Figure 1) is the part that is most exposed to the direct proton beam which, with an initial energy of 18 MeV, stops entirely in the loaded enriched water volume, after traversing the target collimator and the two windows (Figures 2 and 3). The consequent release of heat in enriched water (about 160 W/cm², reaching a temperature of about 240°C at 20 bar standard irradiation pressure) causes erosion action processes which lead to damage to the niobium cell internal surface, while the windows, in particular the Havr® one, which is in contact with enriched water, is submitted to direct proton beam interaction as well as erosive water action.

- The presence of these radiocarbon pollutants affects the final yield since they are competitors and/or inhibitors in all its stages of the subsequent FDG synthesis process.

- In this work we focus on the Monte Carlo and experimental characterization of the radionuclidic impurities contained in the proton-irradiated [18O]H₂O originated in the Havr® window. Monte Carlo simulations, as a support to our experimental measurements, allowed an estimation of a list of the main radionuclides produced in the target Havr® window to be compared with experimental measurements.

Target modelling and Monte Carlo simulations

- Monte Carlo calculation have been performed with the PHTS code (Particle and Heavy ion Transport code System) (Sato et al., 2013) coupled with the substructure estimator D-Chain to simultaneously calculate the activity of radionuclides inside the Havr® window exposed to the proton beam as well as its time evolution during bombardment and after the end of beam

- The target cell model (see Figure 4) is composed by niobium body and Havr® window (thickness 25 μm, density 8.3 g/cm³; typical composition (Goodfellow® disks): 42.5% Co, 20% Cr, 13% Ni, 2.8% Ti, 2.4% Mo, 1.6% Mg, 0.2% C, plus residual Fe).