Enhanced production of ^{99}Mo in inverse kinematics heavy ion reactions

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Abstract. The reaction of a ^{100}Mo beam at 12 MeV/nucleon impinging on a ^{4}He gas-cell target was performed. The ^{99}Mo alongside other coproduced isotopes were collected after the gas target on an aluminum catcher foil and their respective radioactivities were measured by offline γ-ray analysis. In this contribution, preliminary experimental results which are used to discuss the possibility of optimal large-scale production conditions of the produced radioisotopes are presented.

1 Introduction

Today, radioisotopes are commonly used in medicine, both in diagnosis and therapy. A novel method for the production of important medical radioisotopes has been developed at the Cyclotron Institute at Texas A&M University. The approach is based on performing the nuclear reaction in inverse kinematics, namely sending a heavy-ion beam of appropriate energy on a light target and isotopes of interest are collected. The production of the theranostic radionuclide ^{67}Cu ($T_{1/2}$ = 62 h) was performed through the reaction of a ^{70}Zn beam at 15 MeV/nucleon with a hydrogen gas target [1]. The ^{67}Cu radionuclide alongside other coproduced isotopes were collected after the gas target on an aluminum catcher foil and their radioactivities were measured by offline γ-ray analysis. The production of additional radioisotopes is also possible by making use of the forward-focused neutrons from the reaction and allowing them to interact with a secondary target. The main requirement to obtain activities appropriate for preclinical studies is the development of high-intensity heavy-ion primary beams.

The next goal was the study of the production routes for the formation of medically interesting ^{99}Mo (used as ^{99}Mo/^{99m}Tc generator) using cyclotron accelerators, as the production in reactors is not enough to cover the world demand [2]. The ^{99m}Tc as a 140 keV γ-ray emitter (I_γ = 89%) with a half-life of $T_{1/2}$ = 6.01 h, is considered to be an ideal radiotracer and it is estimated to be used in approximately 85% of all nuclear medicine diagnostic scans worldwide. The ^{99m}Tc is produced via β-decay from ^{99}Mo ($T_{1/2}$ = 65.94 h). The amount of ^{99}Mo consumed for radiopharmaceuticals increases year by year [3].

The present work reports on the production routes of ^{99}Mo and other medical radioisotopes using inverse-kinematics nuclear reactions.

2 Experimental method

The experiment was performed at the Cyclotron Institute at Texas A&M University. The beam of ^{100}Mo at 12 MeV/nucleon was delivered by the K500 superconducting cyclotron and transported to the target chamber of the Momentum Achromat Recoil Spectrometer (MARS) [4]. The beam impinged on a cryogenic gas cell filled with ^{4}He gas. The experimental setup is schematically shown in Fig. 1. The cryogenic gas cell had a length of 10 cm with 4 μm Havar entrance and exit windows of 19.0 mm diameter. The target temperature was kept at T = 77 K during all the irradiations. The gas-cell target, the aluminum catcher foils and the Faraday cup to monitor the beam current, were set up in the MARS target chamber. The beam current was periodically monitored and was nearly constant (within 15%). The aluminum catcher foil with 127 μm thickness, mounted in a target frame with 12.7 mm diameter hole and positioned after the gas-cell target, was used to collect the produced ^{99}Mo nuclei along with other coproduced nuclides. In order to measure the neutron yield produced during the irradiation, ten neutron detectors were positioned around the chamber. After an irradiation period lasting a few hours (See Table 1), we turned the beam off, then removed the catcher foil from the frame and eventually placed it in front of a 70% efficient HPGe detector. We then measured the foil's gamma activity to determine the production efficiency for ^{99}Mo, as well as that of other coproduced radioisotopes. Also, the produced isotopes could be detected directly with the MARS spectrometer in coincidence with produced neutrons. Three aluminum catcher

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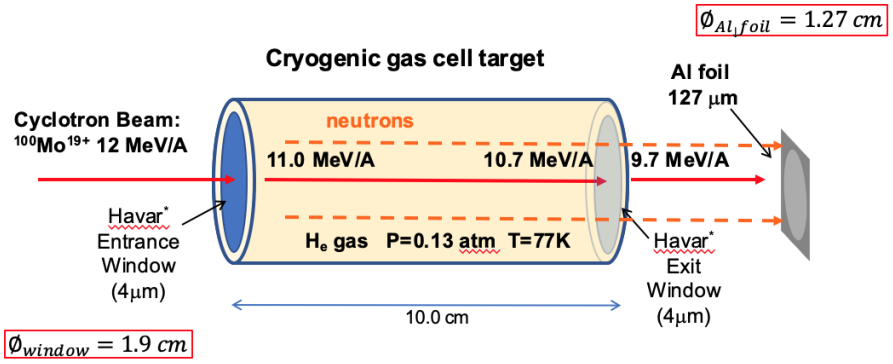

Figure 1. Schematic diagram of the irradiation setup. A ^{100}Mo beam at 12 MeV/nucleon enters the gas cell and interacts with the helium gas. The heavy reaction products, including ^{99}Mo, after exiting the gas cell are implanted in the Al catcher. The energies of the beam are listed as it passes through the entrance window, the gas target and the exit window.

foils (referred to as source 1, source 2, and source 3) were irradiated for different irridiation times, at different beam currents and gas pressures. The irradiation conditions for each foil are shown in Table 1. The beam energy degradation in the gas target and the Mylar windows was determined using LISE++ calculations [5, 6]. We estimated that the beam energy degreaded more when using gas target at higher pressure (source 3). We therefore expected ^{99}Mo and other isotopes of interest to be predominantly produced in source 3.

Table 1. Irradiation conditions for each foil.

Source	Irradiation time (hour)	Beam current (nA)	Gas pressure (Torr)	Target thickness (mg/cm^3)
Source 1	11.6	2.0	102	0.085
Source 2	10.5	7.0	213	0.178
Source 3	7.9	3.5	1008	0.841

A particle-identification plot of the total energy deposited in the Si detectors ΔE–E (55 μm –500 μm) versus the Y-position in the MARS focal plane is shown in Fig. 2. Loci representing different isotopes including mass 99 are clearly visible and well separated.

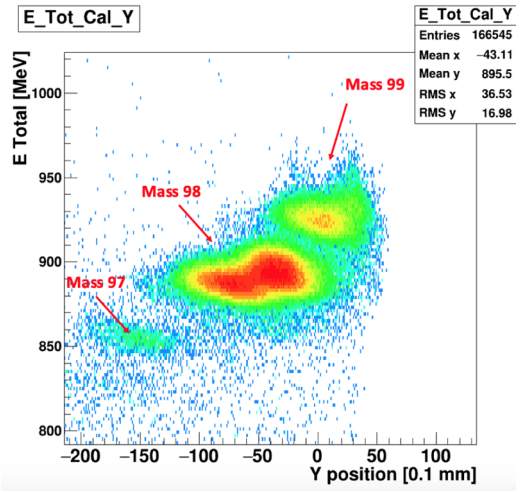

Figure 2. Particle identification spectrum of the total energy deposited in the Si detectors versus the Y-position on the MARS focal plane. Different isotopes including mass 99 are clearly visible and well separated.

3 Data analysis and results

After the irradiations and an appropriate cooling time, the foils were removed, and the gamma activities emitted from the activated foils were measured. The foils were placed at a distance of 50 mm from the end cap of the detector. Under this condition, the dead time of the counting system was around 2–3%, thus avoiding the pile-up effect. The energy resolution of the detector system was 1.0–2.0 keV full width at half maximum (FWHM) for the peaks of interest. The energy calibration was performed using known γ-rays obtained in the spectra. The absolute photopeak efficiencies for the actual source-detector geometry were obtained with the Monte Carlo codes GEANT4 [7] and EGSnrc [8, 9]. The radioactivity levels of the isotopes were determined by the quantification of the photopeaks of the γ-rays taking into account the branching ratios and the absolute efficiencies of the detector. A detailed description of the γ-ray analysis of all the observed γ-ray peaks will be presented in [10]. Room background spectra of good statistics were recorded before the run and carefully subtracted from the spectra acquired with all three sources in order not to affect the uncertainty in determination of the ^{99}Mo yields. Figure 3 displays a background-subtracted γ-decay spectrum for foil 3, obtained during an accumulation period of 16 h, starting 10 h after the end of the irradiation when most of the irrelevant short-lived intense activities of the source (shorter than 1 h) were exhausted. The peaks associated with ^{99}Mo and the most intense impurities are indicated. The most prominent γ peaks are those of ^{99}Mo γ–decay (140.5 keV, 89.4% relative intensity) and

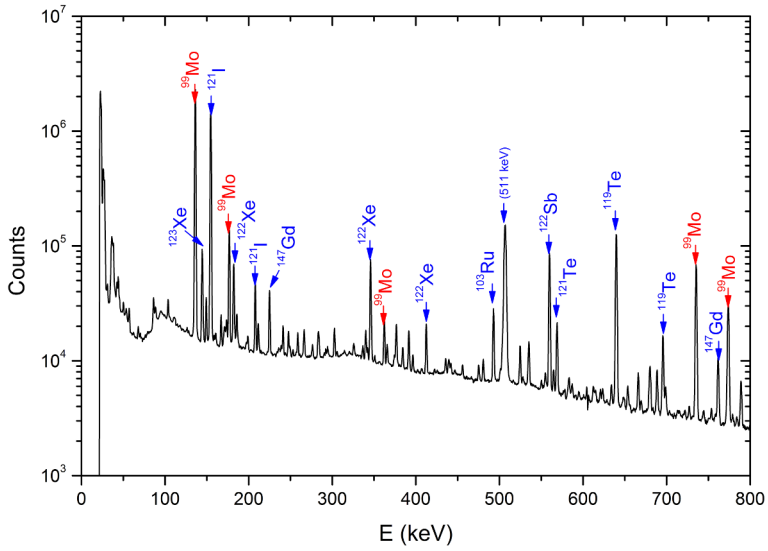

Figure 3. Typical background-subtracted γ-ray spectrum of the Al catcher foil following the interaction of a 12 MeV/nucleon ^{100}Mo beam with the helium gas cell. Different γ-rays coming from the decay of ^{99}Mo along with other radionuclides are indicated.

the other impurities have smaller activities. The reaction presented a breakthrough of ^{99}Mo production. The analysis is underway, the radioactivities for ^{99}Mo and the other impurities are being extracted. The data analysis of radionuclides detected in coincidence with neutrons is underway.

4 Summary

Production routes of ^{99}Mo and other isotopes by means of an inverse kinematics have been demonstrated in this work. This production scheme allows us to use essentially two different targets, a gas and a solid catcher. Furthermore, the neutrons produced in the forward direction could be used to irradiate other targets and produce a variety of radioisotopes of interest. With the neutron detectors, the radioisotope production could also be exploited in coincidence with heavier fragments measured in the MARS detector. We are currently working on the extraction of the relevant cross-sections to compare to data if available and to theoretical models. The present data call for further investigations and the findings will be discussed in the forthcoming papers.

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