Cyclotron production of the ^{105}Rh radionuclide from natural palladium

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1. Introduction

Medical radionuclides used in nuclear medicine are categorized into two principal types: the diagnostics and the therapeutics. The radionuclides that emit gamma (γ) and positron (β^+) with favorable physiochemical characteristics are used in diagnostic purposes. On the other hand, the radionuclides that have the emission of negatron (β^-), alpha (α), electron capture (EC) or isomeric transition (IT) decay leading to the emission of auger electrons with favorable physiochemical characteristics are widely used in therapeutic purposes.

Clinical radionuclide therapy has so far been exclusively practiced using β^- emitting radionuclides. Based on the various physical and chemical properties (half-life, average energy, intensity, mean range etc.), the β^- emitting radionuclides again classified into three sub groups: radionuclides frequently used in clinical purposes, radionuclides used in pre-clinical studies, and radionuclides of potential candidates that have yet to be investigated [1]. The radionuclide ^{105}Rh is considered as a low energy beta (β^-) emitter, and used in pre-clinical studies. It is a good candidate for giving therapy in small tumors (d ≅ 1-2 mm) [2] due to its physical behaviors ($T_{1/2}$ =1.47 days, E_{β^-} = 566 keV, I_{β^-} = 75%, weighted average of beta energy =179.4 keV, and maximum range in soft tissue = 0.89 mm). Beside this, Rhodium complexes are kinetically inert and are expected to be very stable *in vivo* [3]. Currently, large amount and high specific activity ^{105}Rh radionuclide is produced by nuclear reactor using an enriched ^{104}Ru target through the indirect ^{104}Ru(n,γ)^{105}Ru→^{105}Rh process. But, an unsolved problem with current production methods is the high level of Ru impurity associated with the radiochemical separation method used [1]. It is also possible to obtain very large quantities of ^{105}Rh as a fission product, if required. However, alternatively this ^{105}Rh radionuclide can be produced in no carrier added (NCA) form by using medium energy cyclotron through the proton and/or light-charged particle irradiations on palladium target.

2. Experimental

The irradiation technique, the activity determination and the data evaluation procedures were similar to our previous works [4]. Some important features relevant to this work are discussed as follows. The well established stacked-foil activation technique combined with a high-resolution γ-ray spectrometer was employed to determine the excitation function of the ^{nat}Pd(p,x)^{105}Rh reaction. A high purity (>99.99%) and 50 μm thick Pd foil with a natural isotopic composition was used as the target for the irradiation. Several foils of copper (100 μm thick), and aluminum (100 μm thick) together with the Pd target foils were assembled in a stack. The Cu and the Al foils were used to monitor the beam intensity and to degrade the beam energy, respectively. The stacked-foils were irradiated for 60 minutes by proton energy of 42.1 MeV with a beam current of about 100 nA from an external beam line of the MC-50 cyclotron at the KIRAMS. After the irradiations and an appropriate cooling time, the induced gamma activities of the activated foils were measured by using the γ-ray spectrometry. The spectrum analysis was done using the Gamma Vision 5.0 (EG&G Ortec) program. The photo peak efficiency curve of the gamma spectrometer was calibrated with a set of standard point sources. The proton beam intensity was determined by using the monitor reaction ^{nat}Cu(p,x)^{62}Zn with known cross sections from the ref. [5]. The proton energy degradation along the stacked foils was calculated by using the computer program SRIM-2003 [6]. The activation cross-sections for the ^{nat}Pd(p,x)^{105}Rh reaction were determined using a well-known activation formula [4]. The decay data of the radioactive products were taken from the NUDAT database [7].

The uncertainty of the proton energy for each point were estimated from the uncertainty of the incident beam energy, the target thickness, and the beam straggling. On the other hand, the uncertainty of cross-sections were estimated using the uncertainty propagation formula by considering the following uncertainties; statistical uncertainty of the γ-ray counting (~10 %), uncertainty in the monitor flux (~7 %), uncertainty in the detector efficiency (~4 %), and so on. The overall uncertainties of the measured cross-sections were in the range of 8-15 %.

3. Theoretical calculations

The excitation functions of the ^{nat}Pd(p,x)^{105}Rh reaction at the proton energies up to 50 MeV were theoretically calculated using the model calculations by the codes TALYS [8] and ALICE-IPPE [9]. In case of TALYS code, the present results were mostly evaluated using the default values of various models, but the very important inputs like optical model parameters, discrete energy levels and level densities of the nuclides involved in the calculations have been

taken care in a proper way during the calculations. Furthermore, the data of the model code ALICE-IPPE were taken from the web site of International Atomic Energy Agency, where it is compiled as MENDL-2P database [9].

4. Results and Discussion

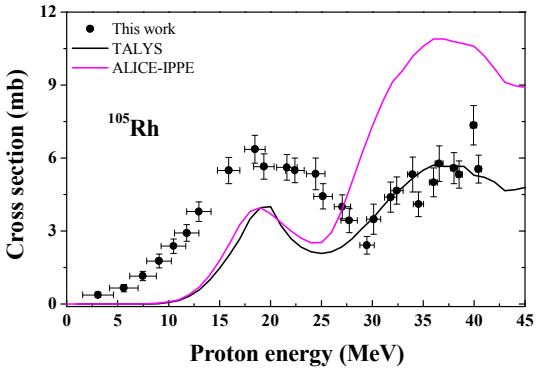

Fig. 1: Excitation function for the ^{105}Rh radionuclide

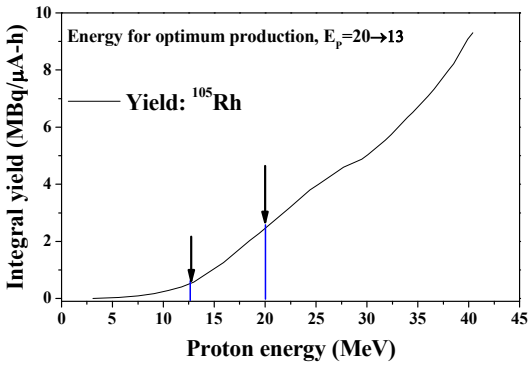

Fig. 2: Integral yield for the ^{105}Rh radionuclide

The long-lived radionuclide ^{105}Rh ($T_{1/2}$=1.47 d) has a short-lived meta-stable state ^{105m}Rh ($T_{1/2}$=40.0 s), which completely decays to the ground state by a IT process before starting our measurement. Therefore, the formation of ^{105}Rh radionuclide follows a cumulative process. The radionuclide ^{105}Rh was identified using its strong and independent gamma line, E_γ = 318.9 keV (I_γ =19.1%). The contributing direct channels for the formation of this radionuclide are the ^{106}Pd(p, 2p) (Q=-9.34 MeV), the ^{108}Pd(p, α) (Q= 3.18 MeV) and the ^{110}Pd(p, 2nα) (Q=-11.78 MeV) reactions. We couldn't compare the present results with any previous measurements due to the lack of available literature data. The measured excitation function of this radionuclide formation is shown in Fig. 1 and compared with the value predicted from the TALYS [8] and the ALICE-IPPE [9] codes. The excitation function predicted by both the model codes TALYS and ALICE-IPPE showed a similar shape to that of the present measurement, and we found a partial agreement with the TALYS prediction above 30 MeV whereas ALICE-IPPE overestimates in this energy region.

5. Conclusions

Production cross sections of the ^{105}Rh radionuclide were measured from the proton irradiations on natural palladium targets using a stacked-foil activation technique in the energy range of 3–40 MeV with an overall uncertainty of 15%. The deduced thick target yield showed that a low energy (<20 MeV) medical cyclotron and highly enriched ^{108,110}Pd targets could be used for a profitable production of ^{105}Rh with a minimum impurity from ^{101m}Rh, even though under above condition the production of ^{101m}Rh impurity is not possible energetically. Above all, the present investigation is the first report on the cyclotron production of carrier free ^{105}Rh radionuclide as an alternative route to the currently used neutron activation process by nuclear reactor.

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