Study of $^{141}Pr(^{14}N,x)$ reactions towards production of medicinally important ^{149}Tb

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Introduction

Medicinally and technologically important radionuclides are generally produced in accelerators using light ion charged particle beams (α, p, ^{3,4}He) [1,2]. However, heavy ion beams from cyclotrons help to produce neutron deficient rare earth lanthanides from non-rare earth elements. It helps simpler radiochemical separation of the radioisotopes of interest from bulk of the target matrix albeit the lower production cross sections of heavy ion induced reactions compared to the light charged particle induced reactions [3]. Some of the rare earth radionuclides are potential candidate for biological applications such as ^{153}Sm,^{149,152,155,161}Tb, ^{177}Lu etc. [3,4]

Among the lanthanides, isotopes of Tb, namely ^{149,152,155,161}Tb (known as 'Swiss knife' family) have got attention as theranostic pair owing to their suitable nuclear characteristics [4]. Among them, the alpha emitting radionuclide ^{149}Tb ($T_{1/2}$= 4.12 h) is a promising candidate for targeted alpha therapy [4-7]. Production of this radionuclide (^{149}Tb) in an adequate quantity and in carrier free form i.e. high radioisotopic purity is a challenge. Production of ^{149}Tb via light ion induced reactions like ^{152}Gd(p,4n), ^{152}Gd(α,7n)^{149}Dy(ε)^{149}Tb and $^{151,\ 153}$Eu(^{3,4}He,xn) have been studied but the limitation of these production methods are requirement of highly enriched target and high energy accelerator though contamination from long-lived Tb isotopes in the final product could not be avoided [4]. Heavy ion induced reactions like ^{142}Nd(^{12}C,5n)^{149}Dy→^{149}Tb have been studied but there is no information on other co-produced Tb radioisotopes in this study [5]. Spallation reactions of ^{nat}Ta(p,x) with 1.0–1.4 GeV proton beam and online mass-separation of isotopes in the ISOLDE (CERN) facility is the only presently avilable method for carrier free production of ^{149}Tb and other Tb isotopes for clinical applications [5].

In the present work, the production of ^{149}Tb via $^{141}Pr(^{14}N,x)$ reactions has been studied. Excitation functions of all the produced radioisotopes are measured using high-resolution gamma ray spectrometry. The results will be compared with theoretical calculations. Thick target yields will also be calculated from the measured cross section data to optimize the production of ^{149}Tb. Radiochemical separation of ^{149}Tb from rest of the reaction products will also be explored.

Experimental

The experiment was performed at VECC, Kolkata using nitrogen beam ($^{14}N^{5+}$) from K 130 cyclotron. Electrodeposited P_2O_3 (600-700 μg/cm^2 thickness) on thin Al foil (12.7 μm) were used as targets. Different stacks each containing 4-5 samples were irradiated to cover the energy range from 80-113 MeV on targets. Targets were irradiated using standard stacked foil activation method [1,2]. The stacks were irradiated with 113 and 109 MeV initial nitrogen beam energies of ~50-60 nA currents for 40 minutes. Energy degradation in the targets of the stack was calculated using SRIM 2013 code. After irradiation, each target was packed in a small polythene pouch and counted several times using HPGe detector system at our Radiochemistry Lab. Energy and efficiency calibration of the detector was done using standard ^{152}Eu source.

Results and discussion

Activation cross sections of all the produced radioisotopes from $^{141}Pr(^{14}N,x)$ reactions were measured using their

characteristics decay gamma rays using standard activation equation [1,2]. A gamma ray spectrum of the 108.5 MeV nitrogen beam irradiated Pr_2O_3 target is shown in Fig. 1.

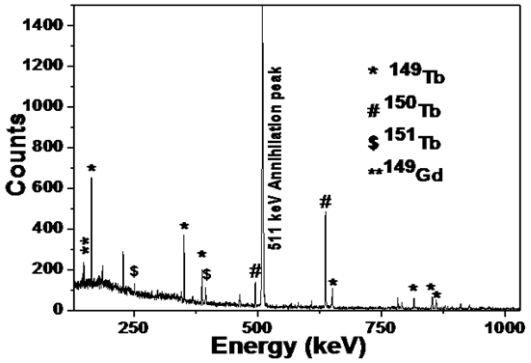

Fig. 1 Gamma ray spectrum of 108.5 MeV nitrogen beam irradiated Pr_2O_3 target, acquired for 15 minutes after 4 h cooling time. Origin of the gamma ray peaks with respect to different isotopes of Tb are marked.

Different isotopes of Tb, $^{148-152}$Tb were identified from the measured spectra. Nuclear spectroscopic data of the studied radionuclides are given in Table 1 [6,7].

Table 1: Nuclear spectroscopic data of the studied radionuclides from ^{141}Pr(^{14}N,x) reactions

Nuclides	**$T_{1/2}$**	**Decay Mode (%)**	**E_γ (keV)**	**I_γ (%)**
^{148}Tb	60 (1) m	EC (100)	784.43	84
^{149}Tb	4.12 (3) h	EC (83.3) α (16.7)	164.98	26.7
^{150}Tb	3.48 (16) h	EC (100) α (<0.05)	638.05	72
^{151}Tb	17.61 (14) h	EC (100) α (0.0095)	251.863	26.3
^{149}Gd	9.28 (10) d	EC (100) α (4.3E-4)	149.73	48

^{149}Tb has two isomers: ^{149g}Tb ($T_{1/2}$= 4.12h, $1/2^+$) and ^{149m}Tb ($T_{1/2}$= 4.17 min, $11/2^-$) and they decay independently to ^{149}Gd ($T_{1/2}$= 9.28 d) without any isomeric transition. ^{149g}Tb ($T_{1/2}$= 4.12 h) is mainly produced via ^{141}Pr(^{14}N,6n)^{149}Dy ($T_{1/2}$=4.23 min, decay via β^++EC (100%)) ^{149}Tb indirectly, whereas direct production via ^{141}Pr(^{14}N,p5n)^{149g}Tb is low [5]. Production cross section of ^{149}Gd was also measured to get total ^{149}Tb produced activity.

Conclusion

Activation cross sections of the $^{149-152}$Tb radionuclides produced from ^{141}Pr(^{14}N,x) reactions were measured from 80 to 113 MeV using standard stacked foil activation method in order to optimize the production of medicinally important ^{149}Tb radionuclide. There are only few measurements available in literature for production of ^{149}Tb using heavy ion induced reactions via ^{141}Nd(^{12}C,x) reactions. To get radioisotopically pure ^{149}Tb, cyclotrons coupled with an isotope separator followed by radiochemical separation is needed. The main purpose of this experiment is to determine activity ratios of potential co-produced radionuclides with respect to ^{149}Tb. All these results will be presented in the symposium.

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