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Theoretical Prediction of the Preferred Production Routes of ^{225}Ac from Proton Induced Reactions on ^{232}Th

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Authors' contributions

This work was carried out in collaboration among all authors. Author ECH designed the study, performed the statistical analysis, wrote the protocol and wrote the first draft of the manuscript. Authors SAJ and OOI managed the analyses of the study. Author HA managed the literature searches. All authors read and approved the final manuscript.

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ABSTRACT

Alpha emitting radionuclides have potential for the therapy of cancers because of their high linear energy transfer, and short range biologic effectiveness. Alpha emitter ^{225}Ac($T_{1/2}$ = 10.0 days) is a potent nuclide for targeted radionuclide therapy. ^{225}Ac excitation functions via ^{232}Th (p,7np)^{225}Th→^{225}Ac, ^{232}Th (p,6n2p)^{225}Ac, ^{232}Th (p,4nα)^{225}Ac, ^{232}Th (p,5n3p)^{225}Th→ ^{225}Ac, and ^{232}Th (p,3nαp)^{225}Ra→^{225}Ac reactions were calculated by Empire 3.2 code up to 200MeV and compared with existing data. No single nuclear level density with a pre-equilibrium model produce results which agree with the existing experimental data all through the energy range. However, a hybrid of the different nuclear level densities with the Hybrid Monte Carlo Simulation (HMS) and the exciton PCROSS pre-equilibrium models at different energy range provide results which are in good agreement with the existing experimental data. Hence the preferred production route for the direct and indirect production of ^{225}Ac has also been suggested.

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1. INTRODUCTION

Despite the present predominant use of beta-emitters in radio immunotherapy trials, the prospective advantages of alpha emitters remain pronounced as acknowledged by clinical and scientific investigations. Alpha – particles, positively charged helium nuclei with shorter range (50-80µm) and higher energy (5,000-8000 KeV) than beta-particles, possess higher Linear Energy Transfer (LET), limited alpha range in tissue [1], improved accessibility and enriched radiochemistry of the emitting nuclides for targeted therapy. As such they offer new prospects in radio-immunotherapy. Over 100 radioisotopes emitting alpha particles are under consideration but quite a number of them decay too quickly to be of therapeutic use [2]. One such isotopes with strong therapeutic potential and fantastic prospect for the cure of malignancy is ^{225}Ac ($T_{1/2}$ = 10 days) which decays by alpha emissions to form ^{221}Fr ($T_{1/2}$ = 4.8 min), ^{217}At ($T_{1/2}$ = 32.3 min) and ^{213}Bi ($T_{1/2}$ = 45.6 min).

It is known that ^{225}Ac is producible by the natural decay of ^{233}U which is under stringent regulation as a weapon material and unavailable in sufficient quantity [3,4] or by the γ-quanta irradiation of ^{226}Ra [5,6] which is also regulated and exceptionally hazardous in gram amounts during irradiation, chemical processing and target recovery. However, some recent measurements have been focused on cross section for high energy production of ^{225}Ac from thorium targets [1,7,8]. Also, recent reports [9,10] which recognized the potentials of this radio nuclide, underscored the necessity for further cross section measurements around several energy regions up to 200 MeV where data are scarce. While available measurements have been documented in the Exchange Format (EXFOR) library, no theoretical calculations have been made available in the evaluated nuclear data library and only limited information is available on optimization of the production of this radio nuclide from thorium target. Therefore, for this study, a reliable modular code, EMPIRE 3.2, has been deployed using adapted optimal model and parameter description for the theoretical prediction of the excitation function of ^{232}Th(p,x) reactions covering the different possible direct and indirect accelerator production routes of ^{225}Ac. The results obtained have been analyzed to propose preferred production routes for the radio nuclide. EMPIRE 3.2 is a modular system of nuclear reaction codes, comprising various nuclear models, and designed for calculations over a broad range of energies and incident particles. The code accounts for the major nuclear reaction mechanisms such as direct, pre-equilibrium and compound nucleus reactions. Direct reactions are described by the optical model, Coupled Channels and Distorted Wave Born Approximation (DWBA). The pre-equilibrium mechanism can be treated by a Multi-step Direct, Multi-step Compound, exciton model (PCROSS) and Hybrid Monte Carlo simulation (HMS). The compound nucleus decay is described by the full featured Hauser-Feshbach model including width fluctuations and the optical model for fission.

2. MATERIALS AND METHODS

Five different exit channels for ^{223}Th(p,x)^{225}Ac reaction have been considered. They include ^{232}Th(p,7np)^{227}Th – ^{225}Ac, ^{232}Th(p,5n3p)^{225}Ra - ^{225}Ac and ^{232}Th(p,3npα)^{225}Ra – ^{225}Ac for the indirect production route. ^{232}Th(p,6n2p) ^{225}Ac and ^{232}Th(p,4nα) ^{225}Ac for the direct production route. Thus an optimal theoretical model and model parameters have been adapted in this study to predict the theoretical excitation functions of proton induced reaction on thorium-232 target leading to the production of ^{225}Ac. The optimal model makes use of the Hauser-Fehsbach (HF) and Hofmann-Richert-Tepel-Weidenmueller (HRTW) versions of the statistical model considering the correlation between the incident and the exit channels in elastic scattering to treat the compound nucleus mechanism [11]. For the Nuclear Level Density contribution to the compound nucleus, a hybrid of the four nuclear level density approaches in EMPIRE 3.2 have been adapted with appropriate tuning of the asymptotic function of the nuclear level densities.

The Hybrid Monte Carlo Simulation (HMS) model and the exciton model PCROSS with appropriate tuning of the mean free path have been adapted to account for the pre-equilibrium mechanism of the optimal model. This optimal model and parameter description predicts the available measured cross section data in good agreement to about 12% Standard Deviation.

3. RESULTS AND DISCUSSION

Table 1 gives the Standard Deviation (SD) analysis of the excitation functions of the different production routes of ^{225}Ac considered is considered. The standard deviation is considered to be sufficiently in good agreement with available measured data. The excitation functions of the five different exit channels have been presented in Figs. 1-3 In each case the cross section shows good trend and agreement with available measured data.

There is very scarce measured data for the production of ^{225}Th from proton bombardment of ^{232}Th. Actinium – 225 is produced through this channel by electron capture. The available measurement of [12] which gives value only at 150 MeV is in excellent agreement with our calculation as shown in Fig. 1. The evaluation of the results of the calculations showed that the threshold located at about 50 MeV, gives a peak characteristics of 215 mb at 85 MeV. This evaluation thus provides theoretical insight into a good model description of this reaction from threshold at 50 MeV to 200 MeV. This indicates that the 7np channel is favored for production from about 50 MeV incident proton source indicating the need for a high energy incident proton beam. The production is however rapidly maximized to peak at 85 MeV indicating an advantage of a short range for the production of ^{225}Th. Fig. 1 further shows that the 7np channel is a more likely route at proton incident energies above 50 MeV and less likely route at proton incident energies below 50 MeV.

The production of ^{225}Ra through the emission of five neutrons and three protons from the proton bombardment of ^{232}Th is one of the indirect routes for obtaining ^{225}Ac through beta decay of ^{225}Ra. The emission of three neutrons, one proton and an alpha particle is also an option to the production of ^{225}Ra. The calculated excitation functions of ^{232}Th(p,5n3p)^{225}Ra – ^{225}Ac and ^{232}Th(p,3npα)^{225}Ra – ^{225}Ac reactions are compared with the existing experimental values in Fig. 2 and are found to be in good agreement with available measured data. [12,13,1] presented the measured production cross

Table 1. Standard deviation analysis of the cross section for the optimal model and parameters with respect to available measured data

The Reactions	Range of the SD for the optimal model description
^{232}Th(p,7np)^{225}Th – ^{225}Ac	SD = 0.09035
^{232}Th(p,5n3p)^{225}Ra - ^{225}Ac	0.008572 ≤ SD ≤ 1.004441
^{232}Th(p,3npα)^{225}Ra – ^{225}Ac	0.005658 ≤ SD ≤ 1.09105
^{232}Th(p,6n2p) ^{225}Ac	0.037985≤ SD ≤ 12.1279
^{232}Th(p,4nα) ^{225}Ac	0.05061 ≤ SD ≤ 8.366227

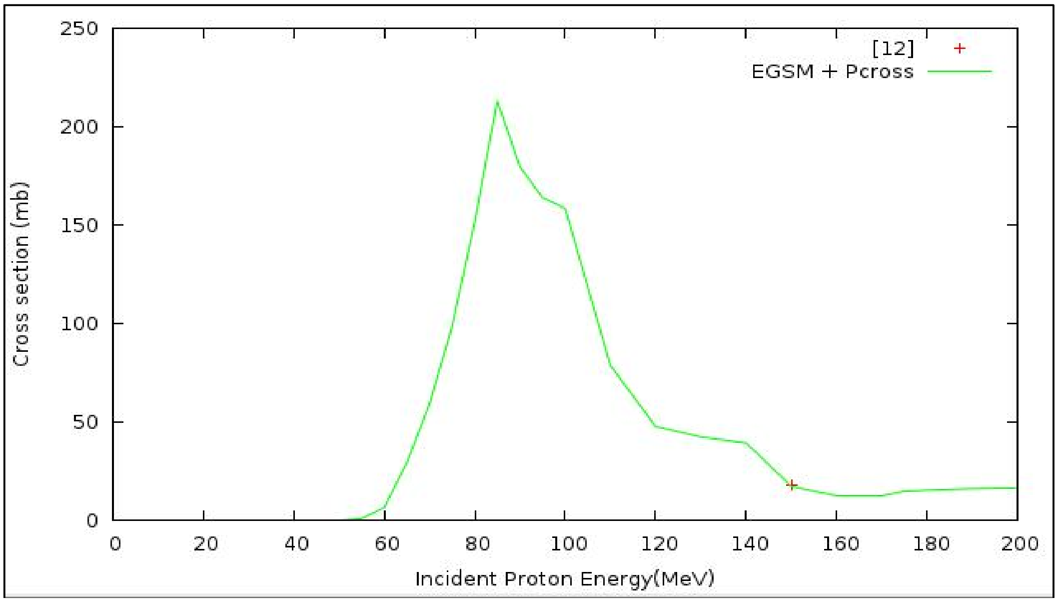

Fig. 1. Excitation function from optimal model used for ^{232}Th (p,7np) ^{225}Th – ^{225}Ac

section of ^{225}Ra without specific prescription of the exit channel and emitted particles. From Fig. 2, it can be observed that the 3npα channel has better agreement compared to the 5n3p channel with respect to available measurements. It is also noted that both channels are favored at proton incident energies below 50 MeV while the 3npα channel is more favored than the 5n3p channels due to its higher cross section up to 50 MeV. From this calculation, the thresholds of these reactions are at 30 MeV for 5n3p and 25 MeV for 3npα. This is in contrast to available measurements obtained only from around 50 MeV. These results can thus serve to provide good model description of cross section from 30 – 50 MeV where there are no measured data for these reactions. Due to its higher peaks throughout the energy range and the other factors indicated the 3npα channel is considered as a preferred route for the indirect production of ^{225}Ac through ^{225}Ra.

The emission of six neutrons and two protons from the proton bombardment of ^{232}Th is one of the possible routes for the direct production of ^{225}Ac. The emission of four neutrons and an alpha particle provides an alternative route for the same reaction. The data from the calculation showed that in order to optimize isotope production the best range of the energy is at 150 to 180 MeV for the 6n2p channel and about 50 – 70 MeV for the 4nα channel.

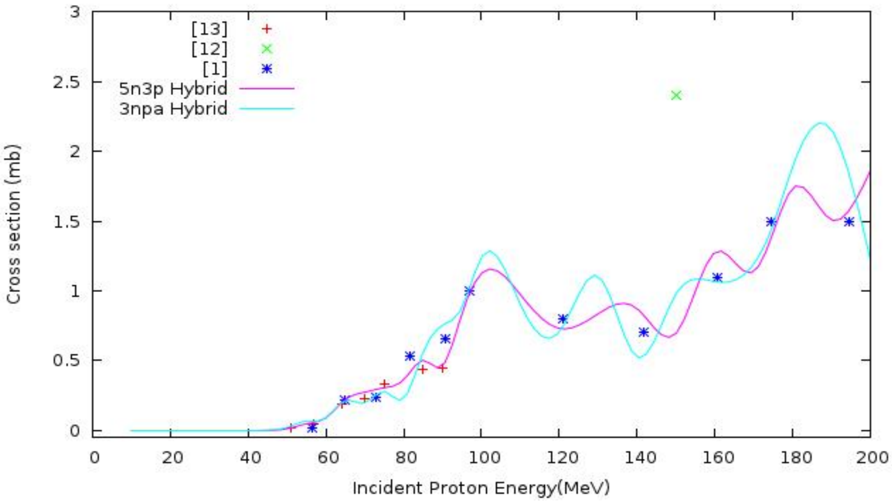

Fig. 2. Comparative plot of the excitation functions of the indirect production routes of ^{225}Ac through ^{225}Ra

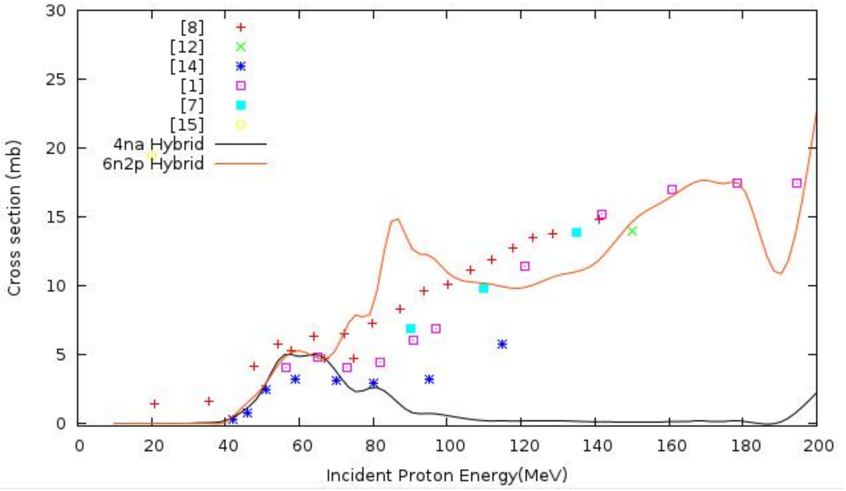

Fig. 3. Comparative plot of the excitation functions of direct production routes of ^{225}Ac

From Fig. 3 the threshold of the reaction can be located at about 25 MeV for both channels but with higher cross section for the 4nα channel. This indicates that the 4nα channel is more favored at lower energy of the proton while the 6n2p channel with higher cross section as the energy increase is favored with more energetic proton sources. However, it is noted that above 80 MeV, the ^{232}Th(p,4nα)^{225}Ac reaction reduces and shows significant poor agreement with the measured values. Furthermore, while the ^{232}Th(p,4nα)^{225}Ac reaction has a lower threshold energy compared with the ^{232}Th(p,6n2p)^{225}Ac reaction, the peak cross sections are generally higher and are observed at lower energies for^{232}Th(p,6n2p)^{225}Ac. It can thus be concluded that the ^{232}Th(p,6n2p)^{225}Ac reaction is energetically more favorable for the direct production of ^{225}Ac.

4. CONCLUSION

Result of the calculations and considerations concluded that for the indirect production of ^{225}Ac the^{232}Th(p,3npα)^{225}Ra – ^{225}Ac reaction route is more energetically favorable for the production of ^{225}Ra (^{225}Ac precursor) than the ^{232}Th(p,5n3p)^{225}Ra – ^{225}Ac reaction production route. Similarly, it is concluded that the ^{232}Th(p,6n2p)^{225}Ac reaction is energetically more favorable for the direct production of ^{225}Ac when compared with the alternate ^{232}Th(p,4nα)^{225}Ac production route.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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