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Actinium-225 Production via different irradiation beams of Thorium-232

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Actinium-225 Production via different irradiation beams of Thorium-232

Naser Burahmah, *University of Tennessee*

Abstract— **Cancer research is developing and expanding on the use of alpha emitting radioisotopes like ^{225}Ac. The demand is increasing so new ways to supply sufficient amounts are needed. Bombarding protons on thorium targets is one method to meet some of the ^{225}Ac demand. National laboratories are researching and developing ways to produce large quantities of pure ^{225}Ac. Deuteron and alpha beams are used to investigate alternate ways to produce ^{225}Ac at accelerator facilities. Simulations were performed using deuteron and alpha beams on a thorium target with energies up to 200 MeV. Deuteron simulations showed great promise to produce larger quantities of ^{225}Ac when compared to production via protons at the same energy per nucleon.**

I. INTRODUCTION

There is interest from the medical community in the use alpha-emitting radioisotopes for therapeutic treatment. One of the techniques used in the nuclear medicine field is Targeted Alpha Therapy (TAT). This new technique shows a promising method for treating cancer and other diseases. The high linear energy transfer of alpha-emitting isotopes in human tissue can kill tumor cells while minimizing the damage of nearby healthy cells. ^{225}Ac ($t_{1/2}$ = 9.92d) and its daughter ^{213}Bi ($t_{1/2}$ = 45.59m) are some of the candidates for TAT. The Department of Energy and other institutions are researching and developing methods for producing large quantities of ^{225}Ac. The research presented here is a continuation of previous work on proton bombardment of Th targets [1]. We are investigating the use of other beams (Deuteron, Alpha) to produce ^{225}Ac. The Monte Carlo simulation code PHITS is used to predict cross sections of ^{225}Ac, ^{226}Ac, ^{227}Ac, ^{227}Th, ^{228}Th and other actinides.

II. ACTINIUM PRODUCTION

The current method to produce ^{225}Ac is by milking it from the decay of long-lived ^{229}Th ($t_{1/2}$ = 7880 y). The supply of ^{229}Th is extracted from the fissile material ^{233}U generated by nuclear reactors or in weapon production. The decay chain of ^{233}U is presented in Fig. 1. The three suppliers of ^{229}Th in the world are Oak Ridge National Laboratory (ORNL), United States, Institute of Physics and Power Engineering (IPPE), Russia and the Institute of Transuranium Elements (ITE), Germany, with a total supply of around 1-2 Ci of ^{225}Ac in global production [2]. Recent research shows another way to produce ^{225}Ac is by bombarding high energy proton beams on ^{232}Th targets through spallation reactions and fission [1]. After the irradiation, ^{225}Ac is separated chemically from Actinide isotopes like radium, thorium, protactinium, and fission products. The only problem in the separation process is the presence of ^{226}Ac ($t_{1/2}$ = 29.37 h) and ^{227}Ac ($t_{1/2}$ = 21.8 y) contaminants. Since ^{226}Ac has a short half-life, it has little effect on the use of ^{225}Ac in TAT, but the long-lived ^{227}Ac cannot be separated. The effect of ^{227}Ac and its daughter products are still being researched. Studies are projected to produce around 1 Ci of ^{225}Ac, equaling the global demand. The goal of this work is to find another method to produce large amounts of ^{225}Ac with less contaminated products (^{227}Ac).

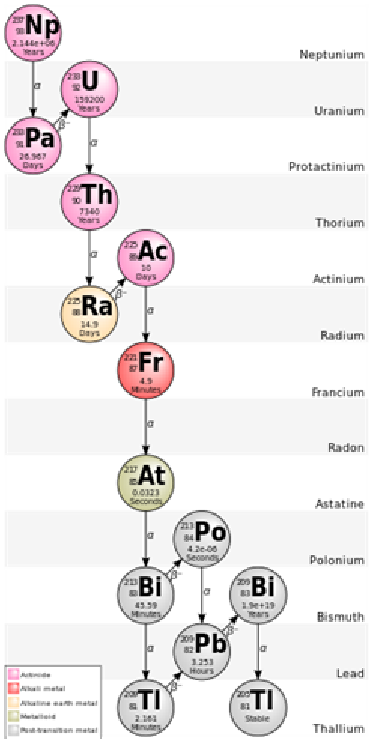

Fig. 1. Uranium-233 decay chain

III. PHITS MONTE CARLO SIMULATIONS

PHITS (Particle and Heavy Ion Transport code system) is a Monte Carlo particle transport simulation code. The code was developed by the Japan Atomic Energy Agency. PHITS transports all particles (neutrons, protons, electrons, nuclei, mesons, nucleons, and photons) over a wide range of energies. The code can be used in facility design, medical physics, radiation protection and geoscience applications. The physics models used in PHITS are shown in Fig. 2. For the simulation, inter-nuclear cascade (INCL4.6) and evaporation (GEM) models were used to calculate the reaction cross-sections of the irradiation of proton, deuteron and alpha beams on ^{232}Th targets in range of energies used in previous studies.

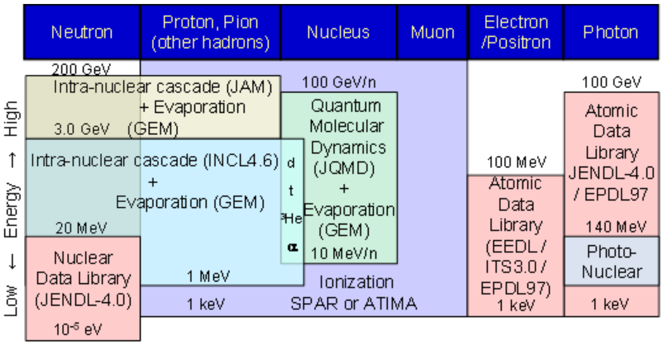

Fig. 2. Physical models in PHITS

In this work, a simple geometry was created, as shown in Fig. 3. The thorium foil target used in actual proton irradiations was modeled as a rectangular solid. The beam was modeled as cylindrical source with 0.5 cm radius targeted on the center of the thorium foil. PHITS *T-Product* tally was used to track the production of nuclides through spallation reactions. In each simulation, 500 million particles were used in the irradiation of the thorium foil. The projectiles used were deuteron and alpha with energies of (76.5, 89.9, 127.8, 152.5, and 192.1 MeV per nucleon). Then, the resulting cross-sections from those projectiles were compared with previous cross-sections for proton beam calculations [1].

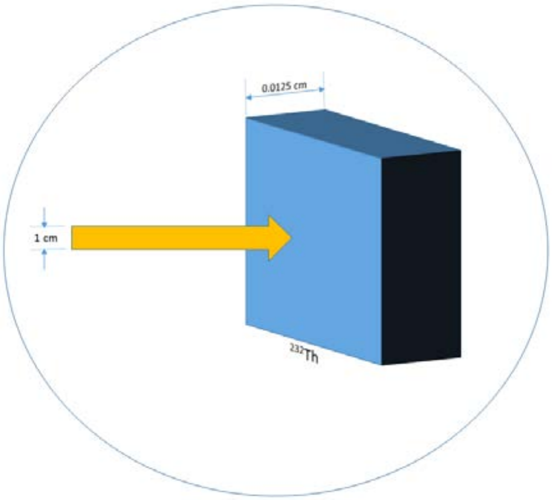

Fig.3. Geometry used in the calculation

PHITS-calculated cross-sections are independent and do not include contributions from the decay of any precursors. PHITS creates text files which contain the number reaction products created in the simulation. A Microsoft Excel Visual Basic Macro was created to process the output file and compute the cross section data for each isotope. The file used to calculate the independent cross-section did so by summing the number of nuclei produced (N_p) and dividing by the total number of particles simulated (N_t) and target density(n_t) in atoms · cm^{-2}, as shown in Equation 1. The uncertainty for the cross-sections was calculated by Poisson statistics as shown in Equation 2, where N is the total number of product nuclei in each simulation.

$$\sigma_t = \frac{N_p}{N_t \cdot n_t} \qquad \textit{Equation 1}$$

$$\sigma_I = \frac{\sqrt{N}}{N} \qquad \textit{Equation 2}$$

IV. RESULTS

Calculated cross-sections of selected isotopes using proton beams are given in Table 1 [1]. Cross sections from deuteron and alpha beams are given in Tables 2 and 3. The ratios of the cross-sections from deuteron and alpha beams to proton beams are given in Tables 4 and 5. The goal was to find a better reaction channel to produce ^{225}Ac. The same geometry was used as reported in the literature to investigate the difference in cross-section of selected isotopes between the different projectile beams. Fig. 4 shows the cross-sections of ^{225}Ac, ^{226}Ac, ^{227}Ac, ^{227}Th, and ^{228}Th using proton, deuteron, and alpha beam. The ratios of ^{227}Ac to ^{225}Ac from the different beams are shown in Table 6.

Table 1: PHITS simulated production cross sections (in mb) using proton beam

Isotopes	Half-life	76.5±0.8 MeV	90±0.4 MeV	127.8±0.5 MeV	152.5±0.5 MeV	192.1±0.5 MeV
^{225}Ac	9.92 d	7.8±0.1	11.8±0.1	26±0.3	28.4±0.3	28.2±0.4
^{226}Ac	29.37 h	7.4±0.1	12.6±0.1	21.8±0.2	23±0.2	22.9±0.3
^{227}Ac	21.77 y	12.2±0.2	17.1±0.1	25.6±0.3	26.2±0.3	25.3±0.4
^{227}Th	18.72 d	37.6±0.3	31.5±0.2	25.6±0.3	22.1±0.2	18.1±0.3
^{228}Th	697.15 d	80.3±0.4	65.9±0.3	51.9±0.4	44.7±0.3	37.2±0.4

Data from Griswold et al., 2016.

Table 2: PHITS simulated production cross sections (in mb) using deuteron beam

Isotopes	Half-life	76.5±0.8 MeV	90±0.4 MeV	127.8±0.5 MeV	152.5±0.5 MeV	192.1±0.5 MeV
^{225}Ac	9.92 d	35.7±0.4	39.9±0.5	40.7±0.5	36.9±0.4	32.7±0.4
^{226}Ac	29.37 h	34.4±0.4	37.2±0.4	35±0.4	31.7±0.4	28.6±0.4
^{227}Ac	21.77 y	46.1±0.5	45.8±0.5	41.4±0.5	38.8±0.5	35±0.4
^{227}Th	18.72 d	28.8±0.4	26±0.4	18.5±0.3	15.4±0.3	13.1±0.3
^{228}Th	697.15 d	65.1±0.6	55.1±0.5	40.6±0.5	35.1±0.4	29.5±0.4

Table 3: PHITS simulated production cross sections (in mb) using alpha beam

Isotopes	Half-life	76.5±0.8 MeV	90±0.4 MeV	127.8±0.5 MeV	152.5±0.5 MeV	192.1±0.5 MeV
^{225}Ac	9.92 d	21.1±0.3	22.3±0.3	24.8±0.4	24.5±0.4	23.9±0.4
^{226}Ac	29.37 h	18.3±0.3	20.1±0.3	22±0.3	22.2±0.3	21.1±0.3
^{227}Ac	21.77 y	23.2±0.3	25.6±0.4	27.3±0.4	27.3±0.4	26.3±0.4
^{227}Th	18.72 d	18.5±0.3	16.9±0.3	14.9±0.3	13.4±0.3	11.9±0.2
^{228}Th	697.15 d	45.5±0.5	55.1±0.5	40.6±0.5	35.1±0.4	29.5±0.4

Table 4: Ratio of Cross Sections Using Deuteron Beam to Proton Beam

Isotopes	Half-life	76.5±0.8 MeV	90±0.4 MeV	127.8±0.5 MeV	152.5±0.5 MeV	192.1±0.5 MeV
^{225}Ac	9.92 d	4.58	3.38	1.57	1.3	1.16
^{226}Ac	29.37 h	4.65	2.95	1.61	1.38	1.25
^{227}Ac	21.77 y	3.78	2.68	1.62	1.48	1.38
^{227}Th	18.72 d	0.77	0.83	0.72	0.7	0.72
^{228}Th	697.15 d	0.81	0.84	0.78	0.79	0.79

Table 5: Ratio of cross sections using alpha beam to proton beam

Isotopes	Half-life	76.5±0.8 MeV	90±0.4 MeV	127.8±0.5 MeV	152.5±0.5 MeV	192.1±0.5 MeV
^{225}Ac	9.92 d	2.71	1.89	0.95	0.86	0.85
^{226}Ac	29.37 h	2.47	1.6	1.01	0.97	0.92
^{227}Ac	21.77 y	1.9	1.5	1.07	1.04	1.04
^{227}Th	18.72 d	0.49	0.54	0.58	0.61	0.66
^{228}Th	697.15 d	0.57	0.65	0.68	0.7	0.73

Table 6: Ratio of cross sections ^{227}Ac to ^{225}Ac

Projectile	76.5±0.8 MeV	90±0.4 MeV	127.8±0.5 MeV	152.5±0.5 MeV	192.1±0.5 MeV
Proton	1.56±0.03	1.45±0.01	0.98±0.02	0.92±0.02	0.93±0.03
Deuteron	1.29±0.02	1.15±0.02	1.02±0.02	1.05±0.02	1.02±0.02
Alpha	1.1±0.03	1.15±0.03	1.1±0.03	1.11±0.03	1.1±0.03

A. Actinium production

Fig. 4 shows the cross-sections of ^{225}Ac from the irradiation of proton, deuteron and alpha beams. For the proton beam irradiation, the cross-section increased from 7.8±0.1 mb at E=77 MeV to 28.4±0.3 mb at E=152.2 MeV. The deuteron beam showed larger production cross-sections compared to proton beam production. At E= 77 MeV/nucleon the deuteron-induced cross-section was 35.7±0.4 mb versus 7.8±0.1 mb for the proton beam. The ratio of deuteron beam cross-sections to the proton beam cross-sections as a function of beam energy per nucleon is shown in Table 4. One of the goals of this work was to discover a way to minimize the production of the contaminate ^{227}Ac which cannot be chemically separated from ^{225}Ac. The presence of ^{227}Ac poses potential issues for nuclear medicine applications where the patient may be subjected to unwanted exposure from ^{227}Ac decay products. At E=77 MeV/nucleon, where the cross-section of ^{225}Ac using deuteron beams was four times the magnitude of the proton beam, the ratio of the ^{227}Ac to ^{225}Ac beam was lower for the deuteron beam than for the proton beam. Table 6 shows the ratio of the ^{227}Ac to ^{225}Ac for different projectiles and for different energies. Alpha beam had a higher cross-section than proton beam at E=77-90 MeV/nucleon. Above those energies, the alpha and proton induced cross-sections were relatively close to each other. The ratio between ^{227}Ac to ^{225}Ac for alpha interactions was almost constant at all energies, which happened to be less than observed with proton beams at E=70- 90 MeV/nucleon. PHITS did not supply information on the reaction pathways to produce ^{225}Ac, so only inclusive cross-sections are reported here. Fig. 5 and Fig. 6 show the cross-sections of ^{226}Ac and ^{227}Ac from the irradiation of proton, deuteron and alpha beams. Validation and verification of these results was not possible due to the lack of experimental data for deuteron and alpha beams. However, from the simulations, the results show great promise using deuteron beams. In all cases studied, deuteron induced cross-sections were larger than alpha and proton induced cross sections.

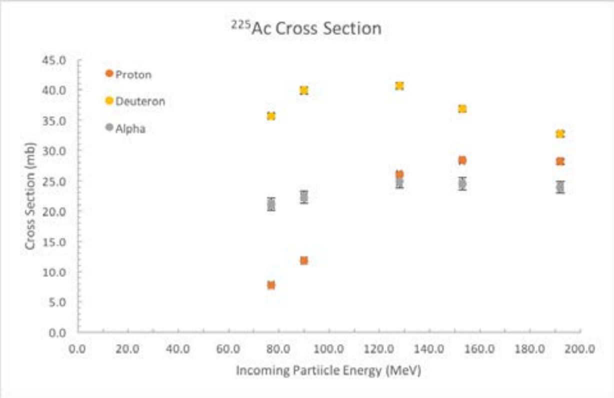

Fig. 4. ^{225}Ac calculated cross-section via different beams

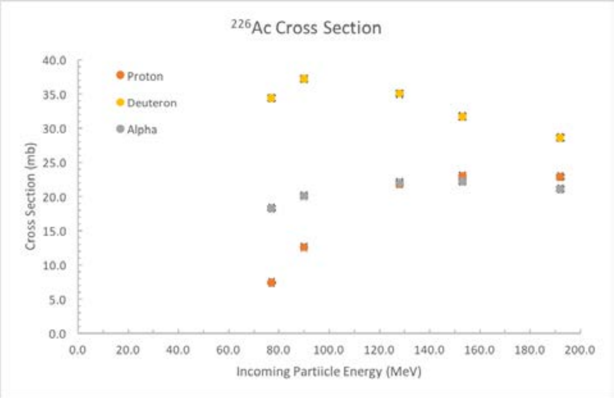

Fig. 5. ^{226}Ac calculated cross-section via different beams

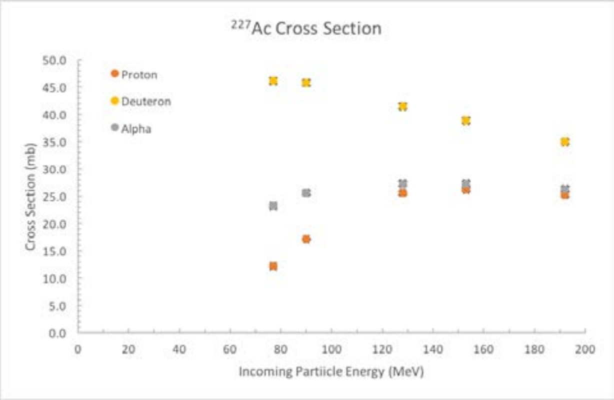

Fig. 6. ^{227}Ac calculated cross-section via different beams

B. Thorium production

Fig. 7 and Fig. 8 shows the cross-sections of ^{227}Th and ^{228}Th from the irradiation of proton, deuteron and alpha beams. Both figures show similar cross-section behavior. In both ^{227}Th and ^{228}Th cases, the proton beam shows larger production of cross-sections compared to deuteron and alpha beam production. At E= 77 MeV/nucleon the data demonstrated the maximum cross-section produced from both ^{227}Th and ^{228}Th for all of the projectiles. Again, validation of these results is not possible due to lack of experimental data for deuteron and alpha beams. In all cases, proton induced cross-sections were larger than deuteron and alpha induced cross sections for both ^{227}Th and ^{228}Th.

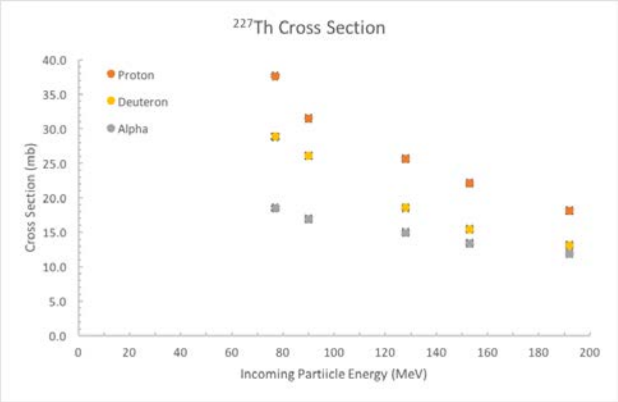

Fig. 7. ^{227}Th calculated cross section via different beams

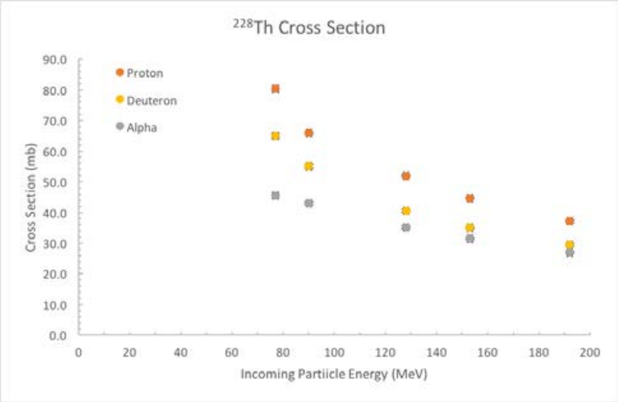

Fig. 8. ^{228}Th calculated cross section via different beams

V. Conclusion

In conclusion, deuteron beam showed great promise for ^{225}Ac production. At E= 77 MeV/nucleon, ^{225}Ac production was a factor of 4 higher for deuteron interactions than for proton-induced interactions. In addition, the ratio of ^{227}Ac to ^{225}Ac using deuteron beams was lower than the ratio from proton beams at the same energy per nucleon. Alpha beams in the lower energies showed better production than proton beams, and as energy increased the alpha-induced production yield decreased to the same level of proton-induced yields. Also, the ratio ^{227}Ac to ^{225}Ac was constant at all energies and was still lower than the ratio in proton beam. Contamination of the ^{227}Ac was lower in both deuteron and alpha beam at lower energies. The results show viable methods of ^{225}Ac production, but validation and confirmation of these results was not available because there was no experimental data to compare to deuteron and alpha beams.

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