Heera Singh, 2025, 13:5
ISSN (Online): 2348-4098
ISSN (Print): 2395-4752

International Journal of Science, Engineering and Technology
An Open Access Journal

Optimization of production routes of various radioisotopes used for industrial applications

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Abstract- Radioisotopes are radioactive isotopes of an element that emits radiations to achieve stability through processes like alpha, beta, and gamma decay. They are used in industry to trace, test and also in several industrial processes and space operations. Radioisotopes are use in RTGs (Radioisotopes thermoelectric generators). RTGs provide electric power using heat from the decay of radioactive isotopes like Plutonium-238 (in the form of plutonium oxide) etc. Works on the principal of Seebeck effect. The selection of fuels for RTGs, there are some criteria that isotopes must have like ability to produce high radiation energy, long half life for regular production of energy, high heat power to mass ratio and cross section. This study is focused mainly on study of cross section area of radioisotopes. The cross section is very important factor, it represents the probability of a nuclear reaction occurring when a particle such as neutron interacts with the nucleus of an atom. This study calculates the cross-sectional area of some radioisotopes with the help of statistical model code EMPIRE 3.2 and compare it with the experimental data available.

Keywords - Radioisotopes, Nuclear cross-section, Radioisotope Thermoelectric Generator (RTG), Plutonium-238, Seebeck effect, EMPIRE 3.2, Nuclear reaction, Decay heat, Half-life, Statistical model, Energy generation, Space power systems.

I. INTRODUCTION

Radioisotopes and Decay Processes

Introduction to Radioisotopes:

In 1896 Henri Becquerel discovered the uranium which leads to discovery of radioactivity. Radioactivity is the phenomenon in which an unstable nuclei decays and loss energy by radiation and emission of particles. [1] This disintegration of nuclei can be done in three ways:

α-decay: The nucleus emits an alpha-particle (Helium nucleus), thus decreasing it's mass number by 4 and atomic number by 2. The reaction concerning the alpha decay is

A X Z A- 4X'Z-2 + 4α2

The alpha-decay plays a major role in RTGs for the production of alpha particles which leads to production of electricity.

β-decay: The nucleus emits a beta-particle (electron/positron), thus changing atomic number by a factor of 1.

The beta-decay involving the emission of electron is β- decay, the concerned reaction is given as

A+1XZ A+1YZ+1 + e- + ύ

The beta-decay involving the emission of positron is decay, the concerned reaction is given as

A+1XZ A+1YZ-1 + e+ + v

γ-decay: In this Process, an unstable nucleus de-excites by spontaneous emission of gamma-ray photon, the reaction is gives as

AXZ AX'Z + γ

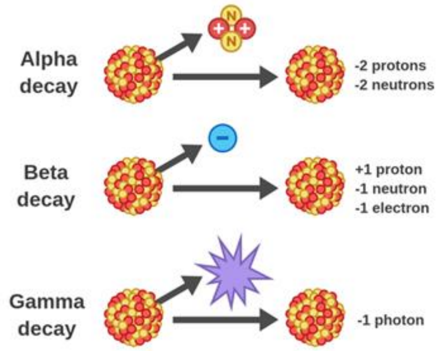

Figure 1.1: A image showing different types of nuclear reaction

Lists of radioisotopes:

RTGs converts heat energy into electrical energy by process of seebeck effect, providing a reliable power source for some space mission like in voyager the heat source is radioisotope fuel Pu-238 in the form of Oxide. While selecting the radioisotopes there was some important aspect like it must have high energy density and long half-life to ensure a stable power output for longer duration of time. Generally, isotopes which have long half-lives, such as Pu-238 half live-87.7years are used in RTGs. The other factor is Radioisotope must have a high heat power to mass ratio. This fulfill the need of heat energy that significant amount of heat generated by radioisotope, useful for compact design. Another factor is radioisotope to have suitable decay process, for RTGs mostly alpha decay process is preferred because of it high energy and low penetrating power. While selecting radioisotope safety plays crucial role the selected isotope must emit low levels of gamma radiation which will help in minimizing the shielding requirement and decrease the radiation exposure risks. Hence selection of radioisotopes for RTGs involves factor like energy density, half-life, decay mode, and safety to ensure reliable and efficient power generation.

Table 1.1 Examples of Radioisotopes with their half-lives, decay process and chemical form.

Isotope	Half-life	Decay Modes	Chemical Form
Am-241	432.2 years	α- decay	Am2O3
Cm-244	18.1 years	α- decay	Cm2O3
Po-210	138 days	α- decay	Po2O3

Uses of radioisotopes:

Radioisotopes plays very important role in modern science and technology. Radioisotopes are unstable due to this nature of their, they undergo radioactive decay that emission of radiation in the form of alpha, beta and gamma rays. This emitted radiation has a wide range of application across many fields such as medicine, agriculture, cardon dating, etc[2]. The radioisotopes of different elements are used for different purpose.

Radioisotope thermoelectric generators-

Radioisotope Plutonium-238 as a alpha emitter used in RTGs to produce heat and then electricity. This process is used in spacecraft power system, nuclear batteries, etc.

Medical tracers-

A chemical is attached with a small amount of radioisotope and then injected into body[3]. Then the process within the body is traced and detected using sensitive gamma camera[5].

Industrial tracer-

Small amount of radioisotope is allowed to flow through the pipe to detect the leakage in the pipe line, then the reading is taken with the help of GM counter.

Radioactive Monitoring-

Smoke detectors also uses weak alpha particle emitter source to cause ionization and then small current to trigger the alarm[4]. Radioisotopes are also used to monitor and control the thickness of paper/plastic/thickness of metal foil.

Radioactive dating-

Cardon dating which uses C-14 isotope of carbon can be used to know how much C-14 is left in once living object.

Seeback effect:

The Seebeck effect is a simple phenomenon in which there is a temperature difference between two different conductors/semiconductors which will produce a voltage difference between the two substances. It was discovered by a German physicist Thomas Seebeck in 1821. He discovered that when two different metal(used copper and bismuth) wires joined at the ends and forms a loop, a voltage is developed is the circuit if both the junctions are kept at different temperature. This pair of metals forming the circuit is called thermocouple. When heat is applied to either of two conductors or semiconductors, the heated electrons flow towards the cooler side. [6] If the couple is connected to a circuit, direct current (DC) flows through the circuit. Seebeck effect produces small voltages around few microvolts but if the temperature difference is large, some of the Seebeck effect devices produces millivolts. Many of these devices can be connected in series to increase the output voltage or together to increase the maximum output current.

RTG uses the Seebeck effect to generate electricity by exploiting the temperature difference between the electric field and the environment through a thermocouple. Plutonium-238 is often used as radioactive material. It decays and produces the heat and this heat produces the electricity[7].

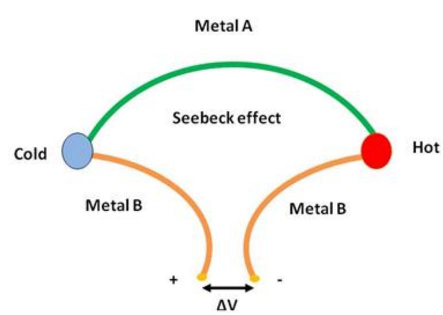

Figure 1.2: Showing the Seebeck effect

Nuclear Cross Section-

It's Interaction between neutron and target nucleus. The standard unit for measuring the cross section is the barn. The larger the neutron cross section[8], the more likely a neutron will react with the nucleus. It is denoted by the symbol σ and the unit is barn, where 1 barn corresponds to an area of 10-28m2. Calculating nuclear cross sections for neutron capture reactions is the primary goal of this study.[9] The neutron cross section, and therefore the probability of a neutron-nucleus interaction depends on

- the target types
- the types of nuclear reaction
- the incident particle energy
- its relative angle between the incident neutron and the target nuclide
- the target nuclide temperature

Computational Tools for Nuclear Cross Section Calculation-

For the computation of nuclear cross sections, computational techniques are essential in nuclear engineering, physics, and other scientific fields. The likelihood that a specific nuclear reaction will take place when a particle interacts with a target nucleus is represented by nuclear cross-sections. Nuclear reactor design, nuclear reaction comprehension in astrophysics, and the development of nuclear medicine applications all depend on these computations. Nuclear cross-section computations are performed using a number of computational techniques and tools like Monte Carlo Simulation, Nuclear Data Libraries, TALYS, EMPIRE and SCALE.

TALYS: (TALYS Nuclear Reaction Program System) is a nuclear reaction simulation code that can calculate cross-sections and other reaction observables for a wide range of projectiles, targets and reaction types. It includes a variety of nuclear models and theoretical approaches, such as the optical model[10].

EMPIRE: (Nuclear Reaction Model Code) is a modular system of codes designed for nuclear reaction simulations, analysis and evaluation. It includes a variety of models and methods for calculating cross-sections, such as Hauser-Feshbach theory and statistical model, as well as various data evaluation tools.

GEANT4: (Geometry and Tracking) is a Monte Carlo code designed for simulating the passage of particles through matter and calculating various quantities, including cross sections. It is widely used in particle physics and medical physics applications and includes a variety of physics models and data libraries.

MCNP: (Monte Carlo N-Particle) is a general-purpose Monte Carlo code that can simulate the transport of particles through matter and calculate various quantities, including reaction cross-sections. It can be used to simulate a wide range of radiation sources and geometries and includes a variety of nuclear data libraries.

SCALE: (Standardized Completer Analysis for Licensing Evaluation) is a suite of integrated software modules for nuclear analysis and design. It includes a variety of tools for calculating cross-sections, such as the TSUNAMI and ORIGEN modules for reactor analysis, and the KENO Monte Carlo code for criticality safety analysis.

II. LITERATURE REVIEW

Lee, Boon Q's Atomic radiations in the decay of medical radioisotopes, in this paper radioactive decay process explained as the unstable atoms release energy by the process of radioactive decay and emits radiations the form of alpha, beta and neutrons particles or EM radiations.

These processes have wide range of applications and are based on interactions of radiations passing through given materials. The majority of radioactive substances used for clinical treatment release ionising radiation in the form of beta particles. this paper mostly deals use of radio isotopes in medical industry as well as radioactive decay processes such that it explains when the other atom is left in an excited state when a vacancy develops in the inner electron shell. The mechanisms of the photo-ionization, ion-atom collisions, electron bombardment, electron capture (EC), and internal conversion electron (CE) can all result in the creation of a vacancy of this kind. The only nuclear decay and nuclear structural modification processes are EC and CE [11].

The paper by Ivan Sykora and klaus Froehlich explore more applications of radioisotopes as tracers and aerosols. Anthropogenic radionuclides which are used for atmospheric radioactivity which have gaseous radionuclides such as 85Kr, 37Ar, 133Xe, and a number of others such as 137Cs, 90Sr, 131I, 144Ce and 106Ru.Nuclear bomb explosions and nuclear facilities (reactors and reprocessing plants) are the primary sources of anthropogenic radionuclide generation and atmospheric emission. The dispersion of the most commonly used radioactive substances, such as 222Rn and its long-lived decay products, 210Pb and 7Be, is the main topic of this paper[12].

Since 2009, a European Space Agency-funded programme has been developing radioisotope power systems that use americium-241 as a heat source and thermoelectric generators in Europe is discussed in this study. This document contains a detailed information of the technical programme and design descriptions and evolution of RTG. This paper also Examine the Production and Consolidation of Fuel Based on Americium-241 for RTGs[13]. Radioisotope thermoelectric generators like 238Pu play a role, in producing power by harnessing the energy from their decay process supporting missions, for space exploration. The study delves into the advantages and disadvantages of utilizing fixed RTGs in present space missions. This paper also tell about Energy Production Process at RTG which is given by ZT refers to a parameter that evaluates the effectiveness of a thermoelectric generator, without any parameters. It takes into account both the thermal properties of the generator outlined as follows

$$ZT = S2T\sigma/k \quad (1)$$

Where S is Seebeck coefficient, σ electrical conductivity, T absolute temperature and k thermal conductivity of the thermoelectrical material[14]. Now the shortage of radioisotope fuel [15] When he researched seven isotope he found that small quantities of 250Cf, 244Cm and 243Cm could generate energy, than 238Pu. These particular samples were deemed secure; 209Po, 232 U, 243 Cm, 244 Cm, 249 Cf, 250 Cf and 251Cf. It is noted that among them the isotope 250Cf stands out as the efficient due, to its energy output compared to 238Pu and these isotopes meet safety requirements.

Selecting the fuel for use, in a Radioisotope Thermoelectric Generator (RTG) can be quite challenging. Initially [16]Dr. Bertram Blankes research on RTG development only identified 47 out of around 1,300 radioisotopes that met the required criteria. These characteristics are very important while selecting are capability to produce high voltage, need to manage heat and prevent distortion, longer lifespan for power production and ratio of thermal energy, to mass (or density). [17] In A. K. Misra, "Overview of NASA Program on Development of Radioisotope Power Systems with High Specific Power," it is mentioned that Radioactive contamination and human safety are always concerns when using nuclear processes in the workplace. Although RTGs are designed to operate in remote areas with few people, this concern is unfounded because there are many unanswered questions about what will happen if RTG oil leaks or an accident occurs during RTG deployment. In the worst case scenario, it can cause radiation damage to humans and serious damage to ecosystems.

[18] In J.A. Rumerman, "NASA Historical Data Book Volume VII it mentioned that how graphite blocks are used to make sure we don't have any contamination risks from RTGs, we take certain safety measures. For instance, during NASA's Cassini-Huygens mission to Saturn, they stored the RTG isotope fuel in high-energy graphite blocks and covered it with a layer of iridium metal to reduce the chances of an explosion. These graphite blocks have proven to be safe, as shown by the Apollo 13 landing mishap in 1970. The spacecraft actually dropped its RTG into water when it returned to Earth, but no plutonium contamination was found. So, it's clear that we've taken precautions to prevent any radioactive pollution.

The optical potential model (OPM) is not only a handy tool in nuclear physics but also finds important applications in nuclear medicine. Scientists rely on accurate predictions of nuclear reactions to develop radiopharmaceuticals for use in nuclear medicine, and that's where OPM comes in. It allows them to accurately predict these reactions. By incorporating radioactive isotopes into active molecules, scientists can produce radiopharmaceuticals that are used in medical procedures.

In a study by Ashok Kumar et al., they investigated alpha nuclear optical potential model (OPM) calculations for a wide range of nuclei with energies up to 140 MeV.[19] They also study the responses associated with alpha particles in the input and output channels The methodology they used included volume integrals and schemes to obtain global optical potential values applicable to different nuclei and energies. They paid attention to modifying the energy dependence at low energies to satisfy the dispersion equation. Calculations of the elastic scattering cross section are in good agreement with the data at high energies, but there are some deviations at low energies. It is important to note that this study has limitations. Factors such as core structure can affect the parameters of the optical model and the assumption of a spherical potential may not always be accurate. There are also discrepancies with the data for the reactions (α,n) and $(\alpha, 2n)$ at higher energies.

Empire

Induction:

The mathematics and theory underlying nuclear reactions are complex. It is inefficient to use multiple complex equations across a wide range of energies to produce a single cross section graph [20]. Nuclear reaction simulations software's are used to calculated or solve this mathematics. Empire a nuclear reaction simulation software was developed by M. Herman. Empire has its own interface and customisation can be easily done depending upon your choices. It can simulate the reaction, generate plots and more. EMPIRE is intended to be a general-purpose, flexible, and user's friendly tool for the analysis and evaluation of fundamental nuclear data. You can choose from a wide range of input parameters, combine various theoretical methodologies, and calculate a lot of observables in a single run. Nuclear data assessment is made simpler by the ENDF-6 formatting, file verification, and graphical comparison with experimental data [21].

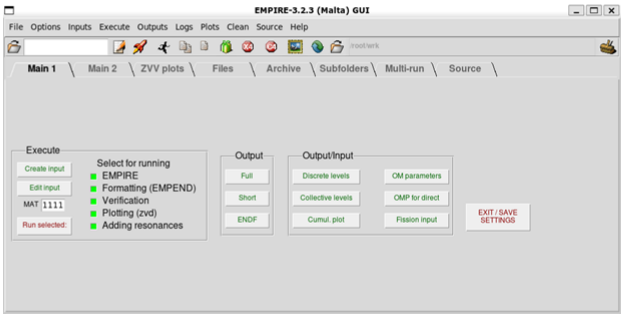

Figure 2.1: The Empire 3.2.3 interface home screen

```
 0.0001              ;INCIDENT ENERGY (IN LAB)
39.   18.           ;TARGET  A , Z
  1.    0.           ;PROJECTILE A, Z
0                    ;NUMBER OF NEUTRONS  TO BE EMITTED
1                    ;NUMBER OF PROTONS   TO BE EMITTED
0                    ;NUMBER OF ALPHAS    TO BE EMITTED
0                    ;NUMBER OF DEUTERONS TO BE EMITTED
0                    ;NUMBER OF TRITONS   TO BE EMITTED
0                    ;NUMBER OF He-3      TO BE EMITTED
0  0. 0.             ; reserved
```

Figure 2.2: A sample Empire input file, describing the reaction 39Ar(n, p)39Cl

Parameters considered for calculation-

a) Incident particle energies: In an EMPIRE input file, we need to provide the range of energy values of incident particle.

b) Nuclear Level densities (LEVDEN): Nuclear level densities are used to measure

reaction cross section values for different values of energies of the incident particles. To describe nuclear level densities, different approaches are used.

c) Optical Model Potential (OMPOT): When a particle is incident on a nucleus, it is scattered, absorbed, and can pass through the nucleus. A similar analogy of behaviour of light can be taken here when it passes through a material of complex refractive index[22]. Quantum mechanics allows us to treat particles as waves and vice versa and thus a complex potential can be approximated that can scatter, transmit or absorbed the particle. This is called the optical model potential (OMPOT). OMP is a very important and useful tool in nuclear physics which is helpful in the study of nuclear reactions and their cross sections.

The form of OMPOT is:

$$U(r) = V(r)\ iW(r) \qquad (2)$$

Where total potential contains both real (V) and imaginary (W) terms same as the scattering of light in a dielectric medium is represented by a complex index of refraction[23].

Methodology of Calculation of cross-sections:

The process outlined involves calculating nuclear cross-sections with alpha particles and target nuclei using the Optical Potential Model (OPM) and the Schrödinger equation. Initially, the OPM is computed to describe their interaction, with parameters often derived from experimental or theoretical data. Subsequently,

the Schrödinger equation is solved to determine the scattering of alpha particles off target nuclei, considering various scattering angles. From these solutions, the cross-section for the desired nuclear reaction is computed, factoring in kinematic considerations and integrating over scattering angles and final states. Analysis and parameter fitting using EMPIRE 3.2 MALTA refine the calculation iteratively for a better description of the reaction. EMPIRE employs mathematical techniques such as partial wave expansion and numerical methods to compute scattering matrix elements, crucial for estimating cross-sections.

This process, combining theory, experimentation, and computation, yields accurate predictions of nuclear reaction cross-sections, offering significant insights into nuclear phenomena and applications [24].

Creating a list of Radioisotopes:

Before creating input files in EMPIRE, a list of isotopes of interest was created.

Table 2.1: list of selected radioisotopes for the nuclear cross section calculations

S.no	Element	Z	A	Half Life	Decay Mode

1.	Americium	95	241	432.2 years	α decay
2.	Strontium	88	38	82.76 days	α decay
3.	Plutonium	239	94	24,100 years	α decay

Nuclear reactions of selected Radioisotopes:

Before giving input to Empire we have to analyze the radioisotopes and their properties like half-life, power density, decay modes etc. Mostly alpha decay is used in RTGs it because alpha decay has high energy output, so they produce effective amount of heat and this heat is converted into electricity. Alpha particles have less penetration power, so risk of damage or exposure to surrounding components and environment reduces. They provide stability most of the alpha particles emitting elements have long half-lives, it ensures a stable and reliable energy output.

This stability is very much needed for space missions. Half-life is also a very important factor to look into specially when we are selecting the radioisotopes, it tells us about the lifetime of reaction before it completely decays. Mostly elements with longer half-life are useful for the RTGs. It also determines the rate at which the radioisotope decays and emits particles. So our reactions are (n,a) type, it is neutron capture followed by alpha decay, when target a neutron towards the nucleus of selected radioisotopes the reaction takes place by the emission of alpha particle.

This reaction mostly seen in nucleosynthesis and nuclear reactors. The three steps in which is reaction can be simplified 1. Neutron capture 2. Formation of excited nucleus 3. Alpha Decay.

The reaction for which the cross section will be calculated are –

Reaction 1:

1n0 + 241Am95 238Np93 + 4He2

Reaction 2:

1n0 + 88Sr38 85Kr36 + 4He2

Reaction 3: 1n0 + 239Pu94 236U92 + 4He2

Results and Discussion

Data Collection from Empire 3.2:

Production of a radioisotope is usually carried out through nuclear reactions and the probability for the production cross-section depends on the sensitivity of the projectile chosen for the reaction. It is expressed in terms of nuclear cross section, so the cross section of selected radioisotopes 241Am, 88Sr, and 239Pu is evaluated using the statistical mode code EMPIRE 3.2 are describe in figures below. The calculated results are compared with the experimental data wherever available as reported in EXPOR.

Reaction of Am-241: The production route of 238Np from 241Am is through neutron-induced nuclear reaction 241Am(n,α)238Np. When a fast-moving neutron hits the nucleus of Am-241 atom which results the absorption of neutron by the nucleus hence as result Np-238 formed along with release of alpha particle. We give the necessary parameter from the reaction to the empire 3.2 and select the energy range and after running the input file we get different values of cross section corresponding to different values of energy.

Then from EXPOR extract experimental data and error and compere the theoretical result with experimental values in a plot. The value of cross section peak is at around 25Mev incident energy of neutron corresponding to around 2.8mb cross section. This is very sacred experimental data and theoretical values matches with available data in EXPOR [25]data in given energy values.

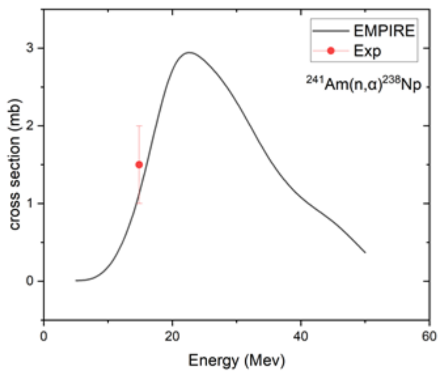

Figure 3.1: The cross section for 241Am(n,α)238Np

Reaction of Sr-88: The production route of 88Sr from 85Kr is through neutron-induced nuclear reaction 88Sr(n,α)85Np. This reaction will also follow the same path as above after analysing the reaction we give the necessary parameter from the reaction to the empire 3.2 and select the energy range and after running the input file we get different values of cross section corresponding to different values of energy.

Then from EXPOR extract experimental data and error and compere the theoretical result with experimental values in a plot. The value of cross section peak is at around 20Mev incident energy of neutron corresponding to around 14mb cross section after this compare experimental data and theoretical values matches with available data in EXPOR [26]data in given energy values.

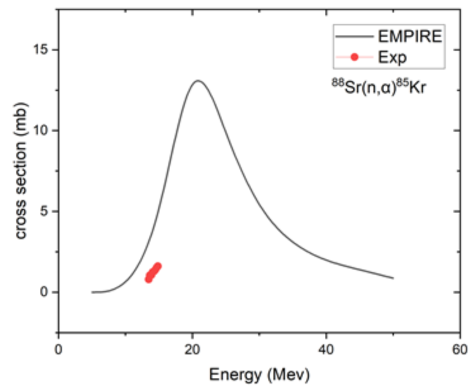

Figure 3.2: The cross section for 88Sr(n,α)85Kr

Reaction of Pu-239: The production route of 239Pu from 235U is through neutron-induced nuclear reaction 239Pu(n,α)235Np. This reaction will also follow the same path as the above reactions after analysing the reaction we give the necessary parameter from the reaction to the empire 3.2 and select the energy range and after running the input file we get different values of cross section corresponding to different values of energy. But for this since there is no experiment data available in EXPOR. But we have completed the theoretical calculations but from the plot the value of cross section peaks at round 24Mev which corresponds to 3mb.

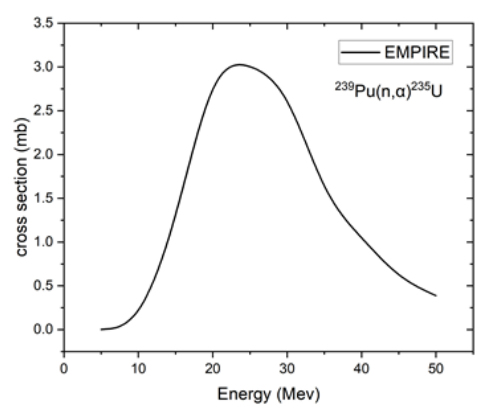

Figure 3.3: The cross section for 239Pu(n,α)235U

III. CONCLUSION

In conclusion, our study involved evaluating the cross sections for the production routes of various radioisotopes essential for Radioisotope Thermoelectric Generators (RTGs). Due to limited experimental data, reliance on theoretical calculations was necessary. However, our calculations showed good agreement with available experimental data in EXPOR. This underscores the reliability of our methods, crucial for optimizing the production routes of these critical radioisotopes. Predicting production yields enhances the efficiency and reliability of RTGs, ensuring their availability for use in space missions, remote locations, and other scenarios requiring long-lasting power sources.

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