Study of proton-induced reaction cross sections for nuclear transmutation of long-lived fission fragments

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1. Introduction

Nuclear power presents a viable solution to reduce greenhouse gas emissions, providing about 10% of the world's electricity. It plays a key role in lowering CO_2 levels and combating climate change, as it is the second-largest source of low-emission energy after hydropower. However, nuclear energy also creates challenges, particularly the waste from fission reactors. These reactors use U^{235} to produce energy, but after they are decommissioned, they leave behind spent nuclear fuel, some of this waste decays quickly while long-lived fission fragments (LLFFs) remain dangerous to human health for thousands of years. Nuclear transmutation is a possible mechanism for reducing the volume and hazard of radioactive waste, aiming to reduce the effective half-lives of LLFFs by making them either short-lived or stable. The purpose of developing a long-lived radionuclide transmutation technology is to provide an alternative option for the geological repository method. Although LLFFs make up only a small portion of reactor spent fuel, their transmutation (or incineration) is essential to lessen the overall waste burden. The transmutation of the LLFFs may be achieved in many ways [1–5]. Transmutation can be carried out either using a nuclear reactor or accelerator. This thesis focuses on providing a strategy for reducing the long-term radiological risks of one such LLFF, ^{126}Sn for which no experimental cross section data exists. The natural half-life of the ^{126}Sn nucleus is 2.18x10^5y. The feasibility of transmutation by bombarding neutrons, protons, and γ-rays should be verified.

The spent fuel in addition to ^{126}Sn contains other Sn isotopes with A=117,118,119,120,122,124. We have to consider not only the transmutation of the radioisotope of concern but also ensure that the other isotopes (stable and short-lived) present do not lead to further long-lived products due to any of the transmutation reactions. In the case of ^{126}Sn, the thermal and fast neutron-capture cross-sections are significantly small ($\sigma^n_{thermal}$=30 mb and σ^n_{fast}=7mb), respectively, and, therefore, not feasible [1]. However, the (p,n) reactions have a cross section of nearly 800 mb for protons on stable isotopes of ^{124}Sn below 15 MeV [6]. Moreover, the residual nuclei formed from (p,n) reactions on Sn isotopes have relatively short half-lives, all under 3 months, therefore, the proton-based transmutation is interesting to pursue.

2. Methodology

The cross section calculations for (p,n) reactions ($\sigma_{(p,n)}$) on Sn stable isotopes were done using the nuclear reaction code TALYS-1.96 [7]. These results were compared with data available from the IAEA Nuclear Data Section. This code includes several nuclear models such as the optical model, compound nucleus, fission, γ-ray strength function, level density (LD), and pre-equilibrium models. The code allows different options to adjust various parameters for each calculation. The optical model potential (OMP) in TALYS code through MOM subroutine, calculates both scattering and absorption cross sections, with the absorption cross sections explaining the formation of a compound nucleus. The statistical model is used to predict the possible decay channels for the absorbed flux. The TALYS calculations have been successfully carried out [8, 9] for various reactions

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*Electronic address: `kavita.churi@physics.mu.ac.in` and applications. In this work, all models and parameters, except for OMP and LD, were kept at their default values. The $\sigma_{(p,n)}$ for stable Sn isotopes were calculated as a function of proton energy, ranging from the reaction threshold up to 20 MeV, using two types of LDs; the back-shifted Fermi gas model (BFM) and the microscopic Hartree-Fock (HF) model based on Skyrme force from Goriely's tables. The Koning-Delaroche (KD) and Jeukenne-Lejeune-Mahaux-Bruyeres (JLMB) OMPs were used for the cross section calculations.

3. Results and Discussion

The main aim is to accurately calculate the $\sigma_{(p,n)}$ and, thereby, the effective half-life of transmutation, $T_{1/2}^{eff}$. The first step is to reproduce the differential and total reaction cross sections (σ_R) for the stable Sn isotopes, as this will ensure that further calculations are in order. In this work, we used the default values of the KD OMP without any modifications, as it is a global parameterization with many parameters. For the case of JLMB OMPs, in order to get reasonable agreement with data for proton on stable Sn isotopes, we had to introduce a renormalization factor for the imaginary part of the central potential, λ_W as 0.85. Further, the calculated $\sigma_{(p,n)}$ calcualted using various combinations of OMPs and LDs for Sn stable isotopes were compared with measured $\sigma_{(p,n)}$ data. Once the models and parameters are fixed, the $\sigma_{(p,n)}$ for ^{126}Sn were predicted. It is found that different combinations of models produced different peak energies, E_{peak}, $\sigma_{(p,n)}$ and $T_{1/2}^{eff}$ for a proton flux of 6.25x10^{14} $s^{-1}cm^{-2}$. The KD-BFM, KD-HF, JLMB-BFM and JLMB-HF gave $\sigma_{(p,n)}$ as 194 mb, 220 mb, 236 mb, and 269 mb, respectively, with E_{peak} ranging from 7.5 MeV to 8.5 MeV. The $T_{1/2}^{eff}$ values were calculated using the simple formula given in Ref. [1]. For the above respective calculations, the $T_{1/2}^{eff}$ were calculated to be and 90 y, 79 y, 74 y, and 65 y, respectively. In comparison, the TENDL-2021 showed a $\sigma_{(p,n)}$ of 180 mb with a longer half-life of of 98 y. In this work, with the same flux, the half-lives for thermal and fast neutrons resulted in in several thousands of years. The $T_{1/2}^{eff}$=65 y [1] was obtained for the JLMB-HF calculations. This is significantly shorter than the natural $T_{1/2}$ of ^{126}Sn. In this thesis, the feasibility of nuclear transmutation of LLFFs, ^{126}Sn using (p,n) reactions have been demonstrated.

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