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Synthesis of neutron-rich transuranic nuclei in fissile spallation targets

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ABSTRACT

A possibility of synthesizing neutron-rich superheavy elements in spallation targets of Accelerator Driven Systems (ADS) is considered. A dedicated software called Nuclide Composition Dynamics (NuCoD) was developed to model the evolution of isotope composition in the targets during a long-time irradiation by intense proton and deuteron beams. Simulation results show that transuranic elements up to ^{249}Bk can be produced in multiple neutron capture reactions in macroscopic quantities. However, the neutron flux achievable in a spallation target is still insufficient to overcome the so-called fermium gap. Further optimization of the target design, in particular, by including moderating material and covering it by a reflector could turn ADS into an alternative source of transuranic elements in addition to nuclear fission reactors.

1. Introduction

Neutrons propagating in a medium induce different types of nuclear reactions depending on their energy. In addition to the elastic scattering, the main reaction types for low-energy neutrons interacting with heavy nuclei are fission and neutron capture, which dominate, respectively, at higher and lower energies with the border line around 1 MeV. Generally, the average number of neutrons captured by a nucleus A during a time interval of Δt is given by the formula:

$$\Delta N = f\sigma_{nA}\Delta t, \tag{1}$$

where σ_{nA} is the cross section of (n,γ) reaction and $f = \langle v\frac{\mathrm{dn}}{\mathrm{d}v}\rangle$ is the neutron flux averaged over the velocity distribution. For example, according to the evaluated nuclear data library ENDF/B-VII [1], $\sigma_{nA} = 1$–3 b for 0.01– 1 MeV neutrons interacting with ^{241}Am and ^{243}Am nuclei. The possibility to capture many neutrons in nuclear explosions was considered already in 60s and 70s, see e.g. Ref. [2], and recently in Refs. [3,4]. In this case the neutron flux is about $3\cdot 10^{30}$ n/(s cm^2), and the explosion time $\Delta t \simeq 1$ μs, therefore:

$$\Delta N_{\mathrm{expl}} = f\Delta t\sigma = 3\cdot 10^{30}\,\frac{\mathrm{n}}{\mathrm{s\ cm^2}}\cdot 10^{-6}\ \mathrm{s}\cdot 10^{-24}\ \mathrm{cm^2} \simeq 3,$$

assuming that $\sigma_{nA} \simeq 1$ b. As shown in Ref. [4], macroscopic quantities of superheavy elements (SHE) with atomic numbers $Z > 100$ located on the Island of Stability can be produced in multiple nuclear explosions.

In ordinary nuclear fission reactors the average neutron flux is typically below 10^{15} n/(s cm^2), and the nuclei up to fermium ($Z = 100$) can be produced there [5]. In this paper we consider the possibility of producing neutron-rich transuranic (TRU) nuclei in Accelerator Driven Systems (ADS) with fissile spallation targets made of americium ($Z = 95$). In this case the average neutron flux could be as large as $3\cdot 10^{16}$ n/(s cm^2) [6]. Since the (n,γ) reaction is only efficient at relatively low energies, $E_n < 1$ MeV, the flux of such neutrons is about twice as low. However, the target can be irradiated by a proton or deuteron beam for a long time, e.g., for one year. Then taking $\sigma_{nA} = 2$ b, one obtains:

$$\Delta N_{\mathrm{ADS}} \simeq 3\cdot 10^{16}\,\frac{\mathrm{n}}{\mathrm{s\ cm^2}}\cdot 3\cdot 10^{7}\ \mathrm{s}\cdot 2\cdot 10^{-24}\ \mathrm{cm^2} \simeq 2$$

The distribution of nuclides in the number of captured neutrons follows a Poisson distribution, providing that all neutron capture cross sections are equal and all other reactions are neglected, see, e.g. [4]. With a simple-minded correction for losses due to fission it can be written as:

$$P_n(\bar{n}) = \frac{\bar{n}^n e^{-\bar{n}}}{n!}(1 - q_{\mathrm{fission}})^n, \tag{2}$$

where $q_{\mathrm{fission}} \simeq \frac{1}{2}$ is the probability of fission in each step. With $\bar{n} = 2$ and $q_{\mathrm{fission}} = \frac{1}{2}$ the probability of capture of 20 neutrons, to produce

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isotopes with atomic mass number $A + 20$, can be estimated as $6 \cdot 10^{-20}$. For a small Am target with its mass of about 40 kg the number of nuclei in the target is $\sim 10^{26}$, therefore, $\sim 10^{6}$ nuclei with mass A + 20 can be synthesized after one year of operation. In this simple estimation all nuclear decays excluding beta-minus decays were neglected.

Neutrons suitable for the production of heavy neutron-rich transuranic elements should have softer spectrum to increase the efficiency of neutron capture reactions. Therefore, such targets should include some moderating material to slow down neutrons. However, too intensive moderation of neutrons would suppress the neutron breeding in fission reactions. A special investigation is required to find a compromise solution. In this paper we model an Am target equipped with a Be reflector by performing detailed Monte Carlo simulations of nuclear reactions and using nuclear data files with realistic neutron interaction cross sections.

2. Modelling of fissile spallation targets

2.1. MCADS — Monte Carlo model for Accelerator Driven Systems

In this work all simulations are performed with the Monte Carlo model for Accelerator Driven Systems (MCADS), which is based on the Geant4 toolkit (version 9.4 with patch 01) [7–9]. This model was used previously for modeling neutron production and transport in spallation targets made of tungsten, uranium and americium [6,10]. MCADS is capable to visualize target volumes as well as histories of primary protons and all secondary particles. It has flexible scoring techniques to calculate neutron flux, heat deposition inside the target and leakage of particles from the target. MCADS makes possible to employ several cascade models for the fast initial stage of pA and dA interactions, which are combined with evaporation, multi-fragmentation and Fermi break-up models for the slow de-excitation stage of thermalized residual nuclei.

In our previous work [10] MCADS was validated with three different cascade models: (1) Bertini Cascade and (2) Binary Cascade both coupled with their standard fission-evaporation codes; and (3) Intra-Nuclear Cascade Liège (INCL) coupled with the ABLA model. Calculations were performed for thin and thick targets made of tungsten and uranium irradiated by protons. For thin ^{238}U targets irradiated by 27, 63 and 1000 MeV protons we have analyzed data on the fission cross section, neutron multiplicity and mass distribution of fission products, and the best agreement was obtained for calculations involving INCL/ABLA. In particular, as demonstrated in Ref. [10], MCADS results obtained with INCL/ABLA model are in good agreement with experimental data available for extended tungsten and uranium targets irradiated by protons in the energy range of 400 – 1470 MeV.

Several extensions of the Geant4 source code and additional nuclear data [11] are needed for the description of proton- and neutron-induced reactions and elastic scattering on TRU nuclei [12]. In [13] MCADS with the modified Geant4 was validated with ^{241}Am and ^{243}Am thin targets. A good agreement with proton- and neutron-induced fission cross sections, fission neutron spectra, neutron multiplicities, fragment mass distributions and neutron capture cross sections measured in experiments was demonstrated.

Alternatively, ADS targets can be irradiated by light nuclei instead of protons. As demonstrated by Monte Carlo modeling in [14] by comparing different beam nuclei interacting with an extended ^{238}U target, 1.5 GeV deuterons provide the highest neutron production rate per unit beam energy. This can be explained by an enhanced probability for deuterons to induce nuclear reactions on heavy target nuclei compared to protons, as the total reaction cross section for deuterons is typically by $\sim$ 40—50% larger compared to the cross section for protons. At the same time the ionization energy loss for deuterons, which is responsible for the stopping of beam particles without nuclear reactions, is equal to the energy loss for protons of the same energy per nucleon. As demonstrated by experimental studies of nuclear residues produced by 4.4 GeV deuterons on gold nuclei [15], the ratio of multiplicities of emitted particles for the deuteron- and proton-induced reactions calculated for 3 GeV protons is 1.5 ± 0.2. Following the findings of Refs. [14,15], in the present study we propose deuterons for the irradiation of an $Am(OH)_3$ target to produce neutron-rich TRU nuclei, see Section 4.

One can note that experimental data on deuteron-induced reactions on actinide nuclei are very scarce. This makes difficult a comprehensive validation of MCADS or any other Monte Carlo model for simulations with deuteron beams. Nevertheless, since the deuteron-induced fission of target nuclei is one of the key processes of neutron production, we have performed MCADS simulations to find the fractions of fission events in thin actinide targets irradiated by deuterons. The total deuteron-induced fission cross sections σ_{df} were estimated from these simulations and compared with the respective experimental data for ^{232}Th [16–18] and ^{238}U [16,19,20] nuclei in Fig. 1. In order to illustrate a general trend of the total fission cross section as a function of beam energy the following analytical parametrization of σ_{df} from Ref. [18]:

$$\sigma_{df}(E) = \sigma_0(1 - \exp(-\alpha(E - E_{th})))\left(1 - d\ln\frac{E}{E_0}\right) \quad (3)$$

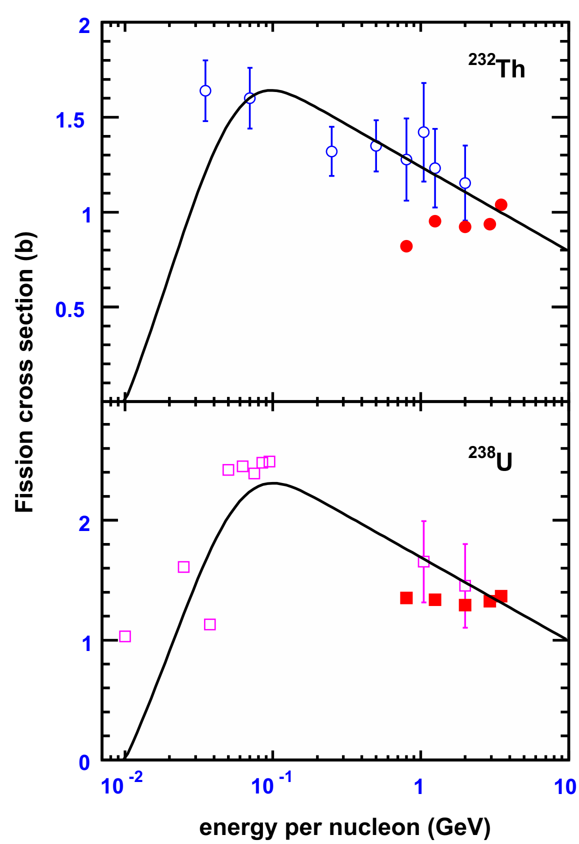

Fig. 1. Total cross section of deuteron-induced fission of ^{232}Th and ^{238}U nuclei as a function of beam energy per nucleon. Experimental data [16–18] for ^{232}Th and [16,19,20] for ^{238}U are shown by open circles and squares, respectively. MCADS results for fission cross sections on ^{232}Th and ^{238}U nuclei are presented by full circles and squares, respectively. Solid lines present the approximation from Ref. [18] which accounts for the decrease of cross section at high deuteron energies, see the text for details.