



ANALYSIS OF AN ADS-FUSION HYBRID TRANSMUTATION SYSTEM USING DIFFERENT TARGETS

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ABSTRACT

This study evaluates the transmutation of nuclear waste in an ADS reactor based on a model proposed by Rubbia and Nifenecker, utilizing different spallation neutron spectra as external sources and reprocessed fuels. The spallation spectra are obtained from a cylindrical target whose geometry was determined in previous studies using a 1 GeV proton beam. The simulated spallation targets are Tungsten (W) and Natural Uranium (U), both solids, as well as Mercury (Hg), Lead (Pb), and Lead-Bismuth Eutectic (LBE) in the liquid state. These fuel assemblies contain an appropriate composition of thorium and reprocessed spent fuels. The JEFF3.3 (Joint Evaluated Fission and Fusion) library, generated for the appropriate temperatures using the NJOY nuclear data processing code, is used for the simulation. The MCNPX 2.6.0 (MCNPX/CINDER) code is employed with the CEM 3.0 (Cascade-Exciton Model) physical model to obtain neutron parameters such as flux at the spallation source surface and the effective multiplication factor (k_{eff}). MonteBurns 2.0 (MCNP5/ORIGEN 2.1) performs a 600 days burnup. The primary objective of this study is to investigate the possibility of transmuting transuranic from spent fuel when subjected to a neutron flux generated by an ADS reactor with an appropriate composition of reprocessed fuel. The results show the system's behavior with the different sources obtained coupled to the simulated ADS reactor. The spallation spectrum of the different targets is inserted into the proposed ADS reactor, and variations in the effective k are verified depending on the burning and the contribution of each target to the creation and destruction of actinides. It is verified that minor actinides were reduced for all targets, which resulted in better performance for the reactor with uranium, tungsten, and mercury targets.

1 INTRODUCTION

Nuclear energy is an important source, with 440 reactors in operation. It currently provides 9% of global energy and can be considered a low-carbon source [1]. Despite its low share, the growth potential is significant, driven by technological advances and the demand for clean energy [2]. However, managing nuclear waste, especially long-lived waste, remains challenging [3]. Transmutation in accelerator-driven subcritical (ADS) reactors has emerged as a promising solution. An ADS uses a beam of accelerated protons to induce spallation reactions in a target, generating neutrons that sustain fission in a subcritical core [4].

The ADS concept gained prominence following Carlo Rubbia's proposal of a fast energy amplifier (EA) system and, consequently, the proposal of a modified concept focusing on transmutation [4, 5, 7].



Unlike conventional reactors, which rely on criticality to sustain chain reactions, ADSs operate in subcritical conditions thanks to an extra source of neutrons. This source is generated by spallation reactions induced in the target by highly energetic protons from an accelerator. Spallation releases many particles, including neutrons, which, when directed toward the subcritical core, ensure the continuity of the chain fissions, acting as an external source of neutrons [6].

The spallation target constitutes the physical and operational interface between the accelerator and the subcritical core. It is the neutron source term for ADS, so it is of great importance to know the number of neutrons emitted by the incident proton (multiplicity), the energy deposited in the target, the angular energy distribution, and the energy distribution of the neutrons, and the distribution of the spallation products.

In a spallation neutron source, the neutron yield from the target directly influences the reactor's thermal power, which is also governed by its degree of subcriticality (k_{eff}). As fuel burnup progresses, k_{eff} decreases, necessitating an increase in the neutron source intensity to maintain constant reactor power. The proton beam parameters must be carefully designed to ensure the target's structural integrity throughout the reactor's operational life [8].

Transmutation is more effective if the minor actinides are fissioned, producing shorter-lived and more treatable fission products. It can occur directly, with a single neutron interaction leading to fission, or indirectly through an initial neutron capture event followed by a second neutron capture leading to fission. A more hardened neutron spectrum in subcritical reactors is important to ensure continuous reactor operation, achieve its goals of waste transmutation and fuel production, and optimize neutron utilization [4].

In this study, the neutron spectra generated by various spallation targets are analyzed, including lead, a eutectic lead-bismuth alloy (LBE), and mercury—each in a molten state—as well as two solid materials: tungsten and natural uranium. The behavior of these materials under spallation was simulated using the MCNPX 2.6.0 code, with the reactor operating at power temperatures. The neutron spectra obtained from these simulations were compared to the default source cited in the MCNP manual for spallation targets [9] and previously used in research at the Department of Nuclear Engineering, UFMG.

It is important to highlight that the simulations assume a reactor operating with liquid coolant temperature (lead) and targets at 600.10 K, while the fuel temperature reaches 1200 K. To compare and analyze the results of different targets, the same system using a reprocessed GANEX fuel spiked with thorium is inserted and burned in an ADS reactor proposed by Rubbia and adapted in a previous work by G. Barros for research in the Nuclear Engineering Department [10, 11, 12] has been evaluated.

The Monteburns code was used for depletion evaluation. Monteburns is a nuclear code that couples the MCNP (Monte Carlo N-Particle) particle transport code with fuel depletion codes such as ORIGEN (Oak Ridge Isotope Generation code). The Department of Nuclear Engineering at UFMG uses Origen2. Monteburns is designed to simulate the evolution of nuclear materials inside a reactor over time, considering the transmutation and decay of radionuclides [10].

2 METHODOLOGY

2.1 Energy and material characteristics

Based on the authors' previous work, this study uses a proton source with an energy of 1.0 GeV, with a parabolic spatial profile, incident on the surface of the spallation targets. In a previous study [reference needed], the ideal thickness of the targets was determined by varying the length of the cylinder between 10 and 80 cm, keeping the diameter constant at 15 cm. Subsequently, the diameter was changed between 10 and 40 cm, keeping the length constant at 50 cm. The neutron yield was calculated for the targets with different diameters and thicknesses, and the ideal size



was suggested based on the neutron yield data and proton range calculations on the targets. Thus, neutron spectra were calculated for targets with optimized dimensions.

In the present study, the target geometry adopted has a height of 50 cm and a diameter of 30 cm. These values were taken from previous studies and proved suitable for simulating the optimized neutron spectrum for an ADS reactor [10]. In previous work, the proton beam's energy varied between 300 MeV and 1.6 GeV, with higher energies increasing the efficiency of neutron production by spallation. However, the maximum neutron production per unit of energy per incident particle seems to occur close to 1 GeV. Therefore, to guarantee beam power greater than 10 MW, proton currents greater than 10 mA are required for 1 GeV proton [5]. The beam has a parabolic profile with an initial diameter of 7 cm and is assumed to be continuous [6]. It is the characteristic, for example, of the proton accelerator of the CIADS (China initiative Accelerator Driven System) project under development [13]

The behavior of the spallation targets was simulated using the MCNPX 2.6.0 code. The source's energy and parabolic profile characteristics were inserted into the SDEF (Source Definition) chart and compared with the standard source applied in previous work. The materials used in this study were chosen for their high atomic number (Z) and thermal properties, suitable for the operating conditions of the ADS reactor, as shown in Table 1.

Tab. 1. Physical characteristics of the materials used, where (a) MP is the molten point, (b) BP is the boiling point, (c) ρ is density, (d) κ is the thermal conductivity [14, 15]

| | Material | Z | MP (K) | BP (K) | ρ (g/cm ³) | κ (Wm ⁻¹ K ⁻¹) |
|---|----------|-------|--------|----------|-----------------------------|--|
| 1 | W | 74 | 3 695 | 5 828.00 | 17.00 | 174.00 |
| 2 | Hg | 80 | 234.32 | 629.88 | 13.53 | 8.30 |
| 3 | Pb | 82 | 600.61 | 2022.00 | 11.35 | 35.00 |
| 4 | LBE | 82/83 | 396.00 | 1938.00 | 10.52 | 22.85 |
| 5 | U | 92 | 1405,3 | 4404.00 | 19.05 | 27.60 |

Solid materials, such as tungsten and natural uranium, stand out for their high thermal and mechanical resistance. In contrast, liquid targets, such as lead, the eutectic lead-bismuth alloy (LBE), and mercury, are preferred for their greater efficiency in dissipating heat and reducing radiation damage. According to data from the literature and technical manuals, materials were selected based on their high densities and thermal properties [16].

2.2 Simulation of neutron production in MCNPX

Neutron production was simulated using the MCNPX 2.6.0 code, with the LAHET model for proton transport and the CEM03.03 model for intranuclear interactions. The shock sections used to describe neutron interactions with the targets were taken from the JEFF 3.3 library. Based on the number of source particles used, the simulations considered uncertainties, with an estimated error limit of 0.2%. The proton beam and its energy and parabolic profile characteristics were defined in the MCNPX SDEF chart, ensuring consistency with previous simulations.

Initially, the proton beam was modeled on the targets, and the number of neutrons generated per proton was obtained. The F4 tally was used to calculate the neutron flux, and the E4 tally was used to determine the energy distribution of the neutrons generated in the spallation reactions. The surface used to measure the neutron flux was positioned around the central region of the subcritical core, ensuring that the volume of interest was equivalent to that of the reactor core.

Once the neutron spectrum had been obtained, the probability distribution was calculated, and the energy range and distribution data were inserted into a new SDEF chart, which was applied to the



ADS reactor model during the burnup simulations with MonteBurns. This new source provided more accurate results for evaluating fuel burnup in the ADS reactor with each specific target. The spectra were normalized to fit the parameters of the modeled ADS reactor spallation source. The target region of the ADS reactor simulated in this work is cylindrical, with a radius of 9 cm, a height of 38 cm, and a total volume of 9669.82 cm³. The target in Rubbia's model is composed of natural lead (Pb) in a liquid state at 600.10 K. This is the default lead target that will be compared with the other five targets simulated in MCNPX. To normalize the neutron spectrum of the spallation targets with volume V (with height H and radius R) so that it fits a new spallation target with volume V' (with height H' and radius R') from the ADS reactor, the following step was taken: volume ratio between the two targets, you can use this ratio to adjust the neutron spectrum. It means that if the original spectrum is $\Phi(E)$, where E is the neutron energy, the normalized spectrum $\Phi'(E)$ will be given by

$$\Phi(E) = \Phi'(E) \frac{V'}{V}$$

This normalization factor adjusts the neutron spectrum to reflect the difference in the volume of the two targets. In addition, the coolant was kept under the same conditions as in the original model, and a specific volume was defined to isolate the neutron source, ensuring that the spatial configuration remained consistent with the reactor's physical constraints. Figure 1 illustrates the geometry of the target and its relationship with the proton beam and neutron production, visually reinforcing the methodology described [6, 7, 9].

2.3 Simulation with MonteBurns

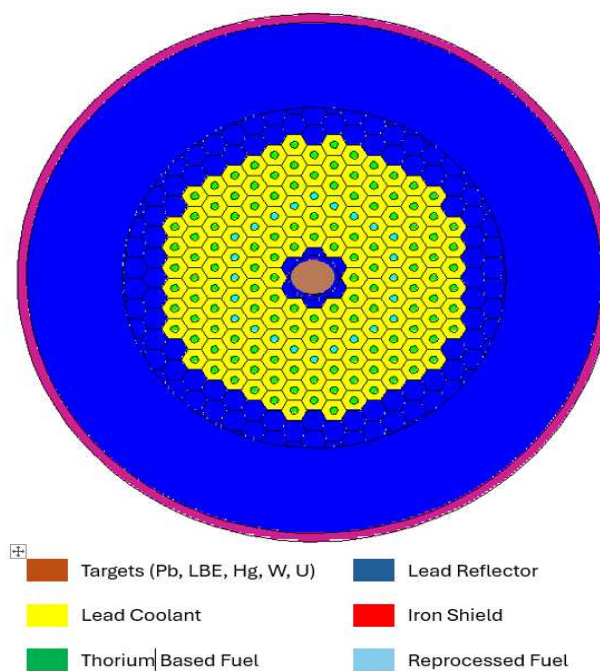


Fig 1 Model of the simulated ADS Reactor containing 132 assemblies of ThO₂ + 17% ²³³UO₂ and 24 assemblies of reprocessed fuel in a hexagonal arrangement.

MonteBurns will be used to simulate the burnup of different targets in the ADS reactor, aiming to analyze the effectiveness of transmutation and identify the most promising target for this strategy. The process involves inputting the target spectra and their respective probabilities into MCNPX's SDEF source card, allowing MonteBurns to model the burnup and production of new isotopes over time. The analysis of simulation results, such as variations in isotopic composition, energy



production, and waste generation, will enable a comparison of each target’s performance in terms of transmutation. The target that shows the highest rate of transmutation of long-lived actinides into products with a shorter half-life or lower radiotoxicity, along with good performance in terms of energy production and waste minimization, will be considered the most suitable for the transmutation strategy in the context of the ADS reactor in question.

The reactor is modeled using its composition $\text{ThO}_2 + 17\% \text{ }^{233}\text{UO}_2$ with 24 spent fuel assemblies subjected to the GANEX process (Group ActiNide EXtraction). This process recovers uranium and transuranics separately, and the TRU (Pu, Am, Cu, and Np) are used to form new fuel diluted with thorium. The configuration of an ADS reactor with an active core composed of 132 assemblies of $\text{ThO}_2 + 17\%$ of $^{233}\text{UO}_2$ and 24 bars of irradiated fuel in a hexagonal arrangement is an adaptation of the Rubbia model first proposed by G. Barros et al. in previous research in the Department of Nuclear Engineering at UFMG [9]

3 RESULTS

The production of neutrons in the target is simulated. From this first simulation, it is possible to obtain the energy distribution of the neutrons generated in the spallation reactions using the tallies F4, which allows us to calculate the neutron flux, and E4, which allows us to assign the energy range of the source.

Fig. 2. shows the characteristics of the neutron spectrum of the materials evaluated (Pb, LBE, Hg, W, and U). The energy of the neutron spectrum was evaluated in a range from 10^{-5} to 10^3 MeV.

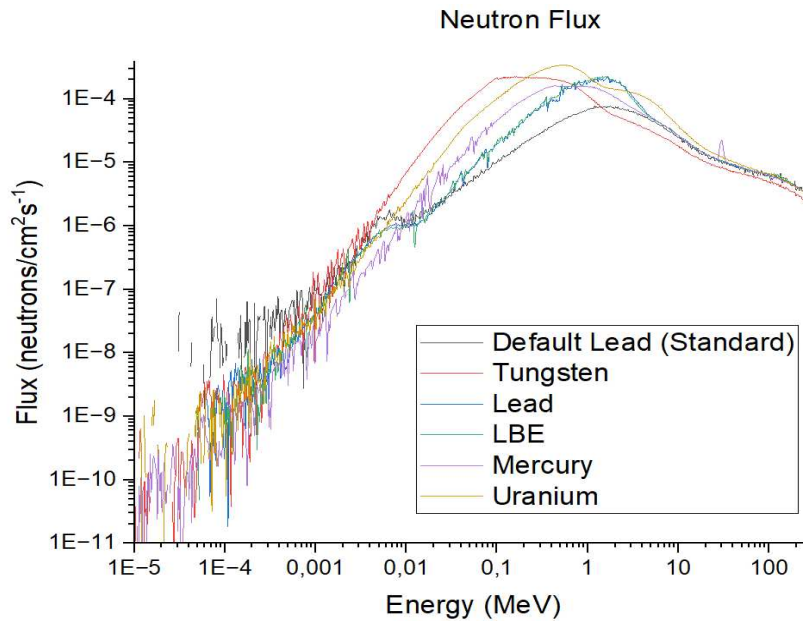


Fig. 2 Comparison of neutron spectra from different targets.

Table 2: Percentage of flux ($\text{neutrons/cm}^2/\text{s}^{-1}$) neutrons per cm^2 per energy interval.

| Material Target | % > 1 MeV | % > 10 MeV |
|-----------------|-----------|------------|
| Tungsten | 17.59 | 3.21 |
| Lead | 58.15 | 6.30 |
| LBE | 57.11 | 6.13 |
| Mercury | 41.12 | 6.23 |



| | | |
|---------|-------|------|
| Uranium | 32.56 | 3.98 |
|---------|-------|------|

Figure 2 represents the neutron flux (n/cm^2s^{-1}) as a function of the energy measured in spallation targets megaelectronvolts (MeV) energy range. It shows that lead and LBE generally have the highest neutron fluxes, followed by mercury, uranium, and tungsten. Lead and LBE have flux peaks at higher energies (approximately 1.6 MeV), resulting in a greater production of high-energy neutrons than other materials. Tungsten peaks at a much lower energy (approximately 0.16 MeV), indicating a less supportable neutron spectrum. Mercury and uranium have slightly lower peak fluxes than lead and LBE.

Lead and LBE have a higher fraction of high-energy neutrons (above 1 MeV), around 58% and 57% (shown in table 2). It confirms that these materials produce a harder neutron spectrum. Lead and LBE are similar in terms of neutron flux and resistance, both producing many high-energy neutrons

The neutron flux increases over -time in systems such as ADS due to the fuel’s characteristics and the spallation target’s behavior.

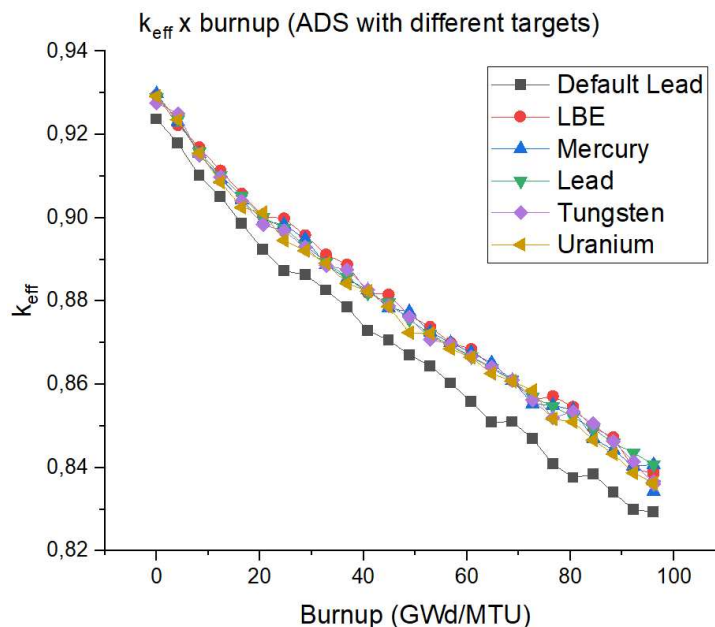


Fig 3. Burnup x k_{eff} for the ADS reactor with different external sources (spallation target) simulated.

As illustrated in Figure 3, it is possible to observe the variation in k_{eff} throughout the burn for ADS reactors, each with a specific spallation target. The reactor with the default source is compared with another ADS with different targets.

It is noted that the ADS reactor containing the LBE spallation source maintains the k_{eff} close to 0.90 even with increased burning, suggesting that k_{eff} must maintain a high value for a longer burning time than the same ADS reactor with other sources. The so-called “default source” has a more irregular behavior than other sources with smoother and better-behaved curves, showing that it is preferable to use a source with the modeled spectrum of the target.

It is possible to observe that in all reactors, there was a net reduction in the total mass of actinides (figure 4). It is a desirable parameter in ADS reactors, aiming to transmute long-lived actinides into less radiotoxic elements or those with shorter half-life. Pu-239 was significantly produced in all cases analyzed. This is an expected result, as Pu-239 is an important fissile nuclear fuel generated by the capture of neutrons by U-238. Pa-231 was produced in very small quantities in



all reactors. Since the protactinium effect is one of the main problems of the Th/U cycle, this low production will not significantly impact the performance of the ADS reactor. Np-239 was also produced in extremely small quantities, close to zero, consistent with Np-239 being a short half-life intermediate in the production of Pu-239.

It is observed that the ADS reactor with the tungsten source had the highest production of Pu-239, accompanied by the greatest percentage reduction in the total mass of actinides (-3.45%). The ADS reactor with mercury source: Pu-239 production was slightly lower, with a percentage reduction in actinides of -3.49%. For the ADS reactor with a lead source, the lowest production of Pu-239 was obtained, and the percentage reduction of actinides was -3.27%. The ADS reactors with uranium and the default source presented the same production of Pu-239 and the same percentage reduction in the mass of actinides of around -3.45%.



Fig 4. Behavior of (a) total actinides, (b) and tree specific isotopes: Pa-231, Np 239 e Pu-239 produced or destroyed in ADS reactor with different targets.

It should be noted that the small percentage of reduction is related to a short burning time, and optimization will depend on maintaining the k_{eff} value in an appropriate range above 0.90 and below 0.98. Optimization of the burning process also depends on optimized arrangements of the reprocessed fuel assemblies inserted into the reactor and the spiked percentual with uranium and/or thorium.

4 CONCLUSIONS

It can be seen that all simulated ADS reactors (i.e., each reactor with a specific spallation target) performed well in the production of Pu-239 and in the reduction of the mass of minor actinides, which is promising for the use of ADS in the transmutation process, in the development of nuclear



systems that can be considered more sustainable and with less environmental impact. It seems that the simulated reactors with tungsten, uranium, and mercury targets performed similarly and slightly more efficiently in the transmutation of actinides. Still, the differences between the targets were relatively small, and optimized simulations with longer burn times will be necessary to verify these results. Optimizing the burn process, including new arrangements of reprocessed fuel assemblies and dilution in uranium and thorium, is a necessary strategy to maximize the production of Pu-239 and the transmutation of actinides while minimizing the production of undesirable products. New arrangements and other reprocessed fuels should be tested in future work.

Future studies are expected to analyze other relevant parameters, such as the production of other minor actinides and fission products, and to perform optimization studies to determine the optimal operating conditions for each target type.

It is important to highlight that the initial study within the limits of the spectra studied and for the parameters adopted demonstrates the potential for transmutation in ADS reactors for nuclear waste management. However, further studies are needed to optimize the process and evaluate the performance of different targets and other relevant parameters.

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