



Depletion and inventory calculation for the new fuel of IEN's Argonauta reactor

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Abstract: This paper presents fuel depletion simulations for the Argonauta research reactor using the MCNP6[®] 1.0 code. The actinide fission yield inventory for the reactor's current U₃O₈ fuel was conducted from 1965 to 2018 using operating parameters obtained from a previous study. Inventories were also generated for hypothetical scenarios involving the reactor's new fuel under development (U₃Si₂). The U₃Si₂ analysis indicated a mass increase for all highlighted actinides across the hypothetical scenarios. Regarding fission product yields, results showed that after 20 years of operation at 1000 W (the new estimated operating power), the mass of all highlighted isotopes was higher than for the same period at 340 W (the current power). These findings are relevant for licensing procedures of the new fuel and for assessing potential accident scenarios, since transitioning to U₃Si₂ fuel and increasing reactor power may impact the isotopic inventory.

Keywords: Argonauta reactor, MCNP, Actinides, Fission products yields.



Cálculo de depleção e inventário do novo combustível do reator Argonauta do IEN

Resumo: Este artigo apresenta simulações de depleção de combustível para o reator de pesquisa Argonauta utilizando o código MCNP6[®] 1.0. O inventário de actínídeos com o combustível atual de U_3O_8 do reator foi simulado de 1965 a 2018 utilizando parâmetros operacionais obtidos em um estudo anterior. Inventários também foram simulados para cenários hipotéticos usando o novo combustível em desenvolvimento (U_3Si_2) do reator. A análise do U_3Si_2 mostrou um aumento de massa para todos os actínídeos destacados nos cenários hipotéticos. Em relação aos produtos de fissão, os resultados mostraram que, ao final de uma operação de 20 anos a 1000 W (nova potência de uso estimada), as massas de todos os isótopos destacados foram maiores do que no mesmo período a 340 W (potência atual). Esses resultados são relevantes para os procedimentos de licenciamento do novo combustível e o estudo de acidentes hipotéticos na instalação, uma vez que a transição para o combustível de U_3Si_2 e o aumento da potência do reator podem resultar em mudanças isotópicas no inventário.

Palavras-chave: Reator Argonauta, MCNP, Actínídeos, Produtos de fissão.

1. INTRODUCTION

Nuclear research reactors are crucial investigative tools, consisting of various materials and geometric configurations that enable nuclear fission through neutrons with specific energy levels. Due to their complexity, safe and effective utilization requires rigorous analysis. Over time, computational methods have evolved from basic simulations to sophisticated tools for reactor analysis. Among them, Monte Carlo (MC)-based codes can simulate physical structures and reactor parameters in detail.

One application of MC-based codes in nuclear science is fuel burnup calculations, which analyze fissile material depletion and generate radionuclide inventories. During reactor operation, atomic densities of various isotopes continuously change, affecting flux, reactivity, and power distribution [1]. The isotopic inventory generated through nuclear reactions is a critical factor due to potential health risks, either through radioactive material release in catastrophic accidents or through long-term fuel storage management.

Radioiodine, noble gases, strontium, and cesium are among the most critical isotopes for accident scenario analysis [2]. Regarding long-term storage, actinides, though produced in smaller quantities than fission products, pose significant disposal challenges due to their long half-lives, often spanning thousands of years. Significant examples include plutonium, neptunium, and americium, which are generated through successive neutron capture in uranium and its byproducts during reactor operation [2]. Therefore, evaluating a reactor core's isotopic composition is essential.

This study aims to perform fuel depletion calculations and inventory analysis for the Argonauta reactor using MCNP6[®] 1.0. Over the years, the Argonauta reactor has served as a key research tool at the Instituto de Engenharia Nuclear (IEN) in experimental reactor physics, neutron physics, non-destructive analysis, neutron activation analysis, radiotracers,

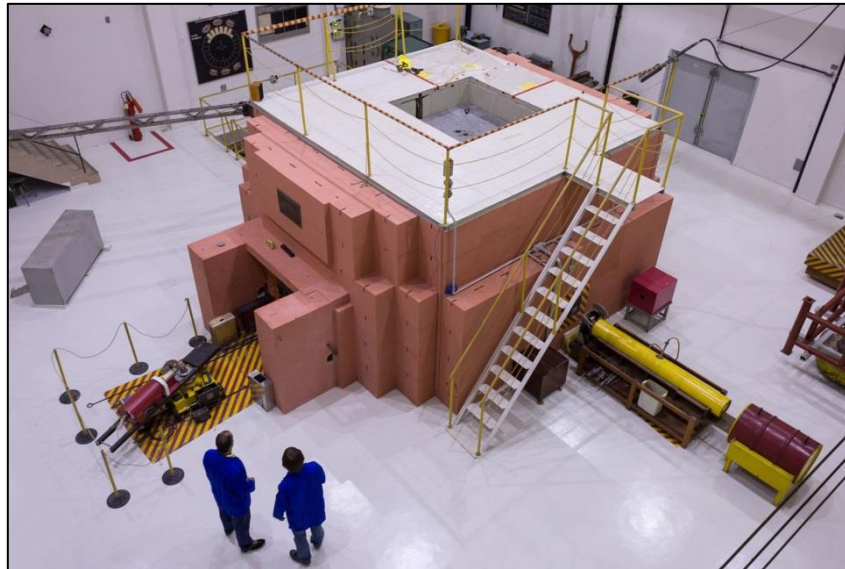
and education. In response to the increasing demand, a study evaluated replacing the current fuel with a modern alternative while maintaining the reactor core's configuration as similar as possible to the existing one. Consequently, inventories for the new U_3Si_2 fuel were calculated under hypothetical operating scenarios. Additionally, inventories for the current U_3O_8 fuel were recalculated based on previous work using a different methodology but under identical conditions.

Although U_3Si_2 fuel offers advantages such as higher thermal conductivity and increased uranium density, early studies suggest it is more chemically reactive than UO fuels. Harp *et al.* (2016) observed the formation of a "layered structure of corrosion products" on U_3Si_2 surfaces after exposure to water at 300°C for 24 hours. The authors emphasized the need for further research while highlighting U_3Si_2 's increased reactivity compared to UO_2 [3]. Replacing the Argonauta reactor's current U_3O_8 fuel with U_3Si_2 necessitates depletion calculations for licensing analysis of the new fuel elements under development.

2. MATERIALS AND METHODS

The Argonauta reactor at IEN was designed by Argonne National Laboratory (USA) under the "Atoms for Peace" program and manufactured and assembled by Brazilian companies. It is a thermal reactor moderated by deionized water and graphite, with these materials also serving as reflectors. Operating at low power, it provides easy access to experimental arrangements for reactor physics and applied nuclear physics research. The facility was officially inaugurated in 1965 and remains the only operational research reactor in Rio de Janeiro [4]. Figure 1 shows a view of the reactor.

Figure 1: IEN's Argonauta reactor perspective view



Source: IEN's website [5].

The MCNP is a general-purpose Monte Carlo transport code developed by Los Alamos National Laboratory (USA) for studying particle interactions over wide energy ranges. The code enables transmutation, activation, and burnup calculations in nuclear reactors. Burnup and inventory calculations involve an iterative probabilistic process requiring reactor operating parameters [6].

The methodology was divided into two parts: simulations using the current fuel based on operating parameters from a previous study (Case Study 1) and inventory calculations using the new fuel under hypothetical operating conditions (Case Study 2). A previously developed model was used as the basis for representing the geometry and composition of the Argonauta reactor [7]. The burn card and relevant parameters were incorporated into the model's input to obtain radionuclide inventories for each case study. The minimum atomic fraction tracked was $1.0\text{E}-15$, using Tier 3 fission product tracking. The simulation included 500 cycles with 500,000 particles per cycle, and an initial keff of 1.03946 (pre-calculated). Simulations were performed on a server equipped with an AMD Ryzen Threadripper 5975WX CPU, using a Linux operating system and parallel processing with 64 cores. Burnup

steps were simplified in the simulation to reduce computational time, as replicating the full operational history of the reactor (e.g., weekly cycles) would be excessively time-consuming and computationally demanding. The simplification involved aggregating weekly operational cycles into annual burnup steps (e.g., 14 days of operation and 351 days of decay per year) to reduce computational cost. Neutron flux and power distribution were assumed constant during each burnup interval.

Case Study 1

In this scenario, the reactor operated with fresh fuel once every seven days for ten months per year, across two power regimes: 170 W for 9,781 hours (1965-1994) and 340 W for 6,504 hours (1995-2018) **Erro! Fonte de referência não encontrada..** To simplify input performance in this work, operation and decay periods were converted into days, resulting in:

- 407.54 days of operation (170 W) followed by 10,177.46 days of decay (0 W),
- 271 days of operation (340 W) followed by 8,124 days of decay (0 W).

The burned material was U_3O_8 fuel, composed of 16.70604% ^{235}U , 68.05964% ^{238}U , and 15.23432% ^{16}O by mass, with total cell volume defined by 116 fuel plates (fuel region only).

2.1. Case Study 2

Simulations were executed using parameters for the new U_3Si_2 fuel, which will be fabricated as a U_3Si_2 -Al dispersion with a density of 2.8 g U/cm³ [8]. Hypothetical burnup scenarios considered reactor operation once every seven days for ten months per year over one, five, ten, and twenty-year periods, at 340 W and 1000 W. Burnup steps alternated between operation and decay:

- 14 days of operation, 351 days of decay (1-year period),
- 69 days of operation, 1,757 days of decay (5-year period),

- 138 days of operation, 3,515 days of decay (10-year period),
- 275 days of operation, 7,030 days of decay (20-year period).

The burned material was U_3Si_2 fuel, composed of 18.46% ^{235}U , 73.84% ^{238}U , and 7.7% ^{14}Si by mass, with total cell volume specified for 132 new fuel plates.

3. RESULTS AND DISCUSSIONS

Table 1 presents the results obtained at the end of the simulation for the mass and activity of these products in Case Study 1. Due to the simplified burn steps, the analysis of the results was restricted to long-lived isotopes.

Table 1: Long-lived actinides 1965-2018

Isotope	Mass (g)	Activity (Ci)
^{232}Th	3.177E-08	3.484E-15
^{234}U	7.397E-05	4.599E-07
^{235}U	2.160E+03	4.669E-03
^{236}U	3.143E-02	2.032E-06
^{238}U	8.801E+03	2.958E-03
^{237}Np	2.255E-05	1.589E-08
^{239}Pu	1.242E-02	7.703E-04
^{240}Pu	3.681E-07	8.352E-08

The results showed some mass differences compared to those from the reference work used for operating parameter comparison **Erro! Fonte de referência não encontrada..** These differences were expected due to variations in the methodologies applied. Tables 2, 3, 4, and 5 present the results obtained at the end of the simulations for the mass and activity of long-lived isotopes in the hypothetical scenarios of Case Study 2, using U_3Si_2 fuel, which was the main objective of this study.

Table 2: 01-year operation long-lived actinides at 340 W and 1000 W

Isotope	340 W		1000 W	
	Mass (g)	Activity (Ci)	Mass (g)	Activity (Ci)
²³² Th	3.168E-11	3.474E-18	9.320E-11	1.022E-17
²³⁴ U	1.275E-06	7.929E-09	1.525E-06	9.479E-09
²³⁵ U	2.080E+03	4.495E-03	2.080E+03	4.495E-03
²³⁶ U	9.199E-04	5.949E-08	2.705E-03	1.750E-07
²³⁸ U	8.318E+03	2.796E-03	8.318E+03	2.796E-03
²³⁷ Np	6.528E-07	4.601E-10	1.921E-06	1.354E-09
²³⁹ Pu	3.389E-04	2.102E-05	9.964E-04	6.180E-05
²⁴⁰ Pu	3.600E-11	8.169E-12	3.113E-10	7.064E-11

Table 3: 05-year operation long-lived actinides at 340 W and 1000 W

Isotope	340 W		1000 W	
	Mass (g)	Activity (Ci)	Mass (g)	Activity (Ci)
²³² Th	6.639E-10	7.280E-17	1.953E-09	2.141E-16
²³⁴ U	6.855E-06	4.262E-08	8.086E-06	5.027E-08
²³⁵ U	2.080E+03	4.495E-03	2.080E+03	4.495E-03
²³⁶ U	4.533E-03	2.932E-07	1.333E-02	8.623E-07
²³⁸ U	8.318E+03	2.796E-03	8.318E+03	2.796E-03
²³⁷ Np	3.221E-06	2.270E-09	9.484E-06	6.684E-09
²³⁹ Pu	1.669E-03	1.035E-04	4.910E-03	3.046E-04
²⁴⁰ Pu	2.228E-10	5.055E-11	1.928E-09	4.376E-10

Table 4: 10-year operation long-lived actinides at 340 W and 1000 W

Isotope	340 W		1000 W	
	Mass (g)	Activity (Ci)	Mass (g)	Activity (Ci)
²³² Th	2.598E-09	2.848E-16	7.639E-09	8.377E-16
²³⁴ U	1.384E-05	8.601E-08	1.629E-05	1.013E-07
²³⁵ U	2.080E+03	4.495E-03	2.079E+03	4.494E-03
²³⁶ U	9.066E-03	5.863E-07	2.667E-02	1.724E-06
²³⁸ U	8.318E+03	2.796E-03	8.318E+03	2.796E-03
²³⁷ Np	6.447E-06	4.544E-09	1.900E-05	1.339E-08
²³⁹ Pu	3.338E-03	2.071E-04	9.819E-03	6.091E-04
²⁴⁰ Pu	2.854E-08	6.476E-09	3.955E-09	8.974E-10

Table 5: 20-year operation long-lived actinides at 340 W and 1000 W

Isotope	340 W		1000 W	
	Mass (g)	Activity (Ci)	Mass (g)	Activity (Ci)
²³² Th	1.023E-08	1.122E-15	3.068E-08	3.365E-15
²³⁴ U	2.778E-05	1.727E-07	3.268E-05	2.032E-07
²³⁵ U	2.079E+03	4.495E-03	2.079E+03	4.494E-03
²³⁶ U	1.807E-02	1.168E-06	5.314E-02	3.436E-06
²³⁸ U	8.318E+03	2.796E-03	8.318E+03	2.796E-03
²³⁷ Np	1.286E-05	9.060E-09	3.797E-05	2.676E-08
²³⁹ Pu	6.649E-03	4.124E-04	1.956E-02	1.213E-03
²⁴⁰ Pu	1.151E-07	2.612E-08	7.973E-09	1.809E-09

Tables 6, 7, 8, and 9 present the results obtained at the end of the simulations for mass and activity of some fission products of interest analyzed in the study of hypothetical accidents.

Table 6: 01-year operation fission products of interest at 340 W and 1000 W

Isotope	340 W		1000 W	
	Mass (g)	Activity (Ci)	Mass (g)	Activity (Ci)
⁸⁵ Kr	4.626E-06	1.817E-03	1.361E-05	5.344E-03
⁹⁰ Sr	1.075E-04	1.519E-02	3.162E-04	4.466E-02
¹²⁹ I	1.461E-05	2.581E-09	4.297E-05	7.591E-09
¹³⁵ Cs	1.871E-04	2.155E-07	5.497E-04	6.333E-07
¹³⁷ Cs	1.757E-04	1.529E-02	5.167E-04	4.497E-02

Table 7: 05-year operation fission products of interest at 340 W and 1000 W

Isotope	340 W		1000 W	
	Mass (g)	Activity (Ci)	Mass (g)	Activity (Ci)
⁸⁵ Kr	1.769E-05	6.947E-03	5.203E-05	2.043E-02
⁹⁰ Sr	4.810E-04	6.794E-02	1.415E-03	1.998E-01
¹²⁹ I	7.318E-05	1.293E-08	2.152E-04	3.802E-08
¹³⁵ Cs	9.219E-04	1.062E-06	2.709E-03	3.121E-06
¹³⁷ Cs	7.907E-04	6.882E-02	2.326E-03	2.024E-01

Table 8: 10-year operation fission products of interest at 340 W and 1000 W

Isotope	340 W		1000 W	
	Mass (g)	Activity (Ci)	Mass (g)	Activity (Ci)
⁸⁵ Kr	2.576E-05	1.012E-02	7.576E-05	2.975E-02
⁹⁰ Sr	8.524E-04	1.204E-01	2.507E-03	3.542E-01
¹²⁹ I	1.466E-04	2.590E-08	4.313E-04	7.619E-08
¹³⁵ Cs	1.844E-03	2.124E-06	5.417E-03	6.242E-06
¹³⁷ Cs	1.412E-03	1.229E-01	4.153E-03	3.614E-01

Table 9: 20-year operation fission products of interest at 340 W and 1000 W

Isotope	340 W		1000 W	
	Mass (g)	Activity (Ci)	Mass (g)	Activity (Ci)
⁸⁵ Kr	2.722E-05	1.069E-02	8.007E-05	3.145E-02
⁹⁰ Sr	1.334E-03	1.885E-01	3.924E-03	5.543E-01
¹²⁹ I	2.925E-04	5.168E-08	8.605E-04	1.520E-07
¹³⁵ Cs	3.674E-03	4.233E-06	1.080E-02	1.244E-05
¹³⁷ Cs	2.243E-03	1.952E-01	6.597E-03	5.742E-01

A mass increase was observed for all actinide isotopes (except for ²³⁵U and ²³⁸U) across scenarios of 1, 5, 10, and 20 years at both 340 W (current power) and 1000 W (new estimated power). At the end of 20 years of operation at 1000 W, the masses of ²³²Th, ²³⁴U, ²³⁶U, ²³⁷Np, and ²³⁹Pu were higher than those at the same period under 340 W. However, the mass of ²⁴⁰Pu at 1000 W was lower than at 340 W. This difference may be attributed to the variation in operating power between the two scenarios and the fission decay rate of ²⁴⁰Pu, as approximately one-third of its atoms undergo fission upon neutron absorption. Regarding the highlighted fission products—⁸⁵Kr, ⁹⁰Sr, ¹²⁹I, ¹³⁵Cs, and ¹³⁷Cs—their masses at the end of 20 years of operation at 1000 W were higher than those at the same period under 340 W.

These findings suggest that the use of U₃Si₂ fuel could result in different actinide accumulation patterns during reactor operation, potentially impacting long-term fuel management, waste disposal strategies, and overall reactor performance. Additionally, the results indicate that increasing reactor power leads to higher fission activity, contributing to

a greater buildup of fission products. This is crucial for safety assessments, as higher fission product yields can influence factors such as radiation levels and cooling requirements.

4. CONCLUSIONS

In this study, simulations were performed using the Argonauta reactor's current fuel and core conditions to obtain an isotopic inventory based on parameters from a previous work (Case Study 1). Additionally, simulations were conducted for hypothetical scenarios using the reactor's new fuel under development, maintaining the same core conditions (Case Study 2). The observed variations in isotope masses in Case Study 1 can be attributed to methodological differences between the two studies, as well as the simplified burn steps used in this work.

In Case Study 2, which was the primary focus of this study, the analysis indicated a mass increase for all highlighted actinides across the hypothetical scenarios. Regarding fission product yields, the results showed that after 20 years of operation at 1000 W, the masses for all highlighted isotopes were higher than those observed at 340 W. These findings suggest that transitioning to U3Si2 fuel and increasing reactor power will lead to significant changes in the isotopic inventory, impacting reactor operation, safety, and regulatory compliance.

Future work suggestions include testing the influence of burn step divisions on depletion performance to refine the accuracy of the simulations.

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CONFLICT OF INTEREST

All authors declare that they have no conflicts of interest.

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