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Evaluation of the mass and activity of Am-241 radionuclide production from different reactors using ORIGEN code

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HIGHLIGHTS

- The mass and activity of Am-241 from the spent fuel of 1000 and 360 MWe reactors are compared.
- Results indicate that the amount of Am increases with the cooling time after removal of SNF.
- The mass of Am-241 from 1000 MWe reactor is about 2.85-8.97 times higher than that of the 360 MWe.
- Irradiated fuel from the 1000 MW reactor can be a suitable source for the extraction of Am.

ABSTRACT

Am-241 is one of the transuranic elements that is used in measuring and controlling the thickness of various industrial materials, diagnosing thyroid disorders, smoke detectors production, designing neutron sources for well logging, etc. Therefore, due to the wide applications of this isotope, research and development for its production, separation, and purification are of great importance. Nowadays, the commercial production of this isotope is done in the United States and Russia. Since the production of Am-241 is carried out from irradiated fuel of power reactors or irradiation targets in research reactors, in this work, the possibility of production and activity of this radionuclide from the irradiated fuel of a typical 1000 MWe and 360 MWe reactors has been investigated using the ORIGEN code. Results indicate the highest production of Am-241 in a 1000 MWe reactor. Therefore, the irradiated fuel of the 1000 MWe reactor can be a suitable source for the extraction of this isotope.

KEYWORDS
Am-241
ORIGEN code
Irradiated fuel
Nuclear power reactors

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1 Introduction

Americium is one of the synthetic actinide elements with the atomic number 95, without any stable isotopes. All isotopes of this element are radioactive. Among the 15 identified isotopes and isomers of americium, the longest half-life is attributed to Am-243. The most important isotopes of americium, Am-241 and Am-243, are significant due to their long half-lives of 433 and 7380 years, respectively. These isotopes have been produced in kilogram quantities with high purity. 242Am, with a half-life of 141 years, can be produced in small amounts by the neutron absorption of Am-241.

Due to the low price, accessibility, and suitable half-life of Am-241, this element is considered as a low-energy gamma-ray source compared to all competing radionuclides (Crandall, 1973; Morss et al., 2011).The production of Am-241 through the (n, γ) interaction with plutonium proceeds as follows:

$$^{239}Pu(n,\gamma)^{240}Pu(n,\gamma)^{241}Pu \xrightarrow{\beta^-} {}^{241}Am \qquad (1)$$

This reaction is presently considered the best method for producing pure Am-241. Am-241 has a long half-life and emits predominant 5.59 keV gamma radiation, which makes it useful for a wide range of industrial measurement applications and for detecting certain thyroid diseases (Seaborg and Loveland, 1991). Additionally, americium is a strong alpha emitter and is used for a number of medical and industrial uses as a portable source of alpha particles, emitting 5.486 MeV energy in 85% of its decays.

The most widespread application of americium is in smoke detectors. One gram of americium dioxide, AmO_2, provides enough active material for over 5000 smoke detec-

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tors. In the 1980s, annual sales of smoke detectors reached 12 million units. This radionuclide, when combined with beryllium, emits 10^{17} neutrons per gram per second. This mixture is used in the design of neutron sources. Am-241 is used as a neutron source in radiography, tomography, and other radiochemical studies. Neutron sources are used in various industries for measurements of density, moisture, and hydrocarbon content. The most extensive use of neutron sources is in well logging operations. The cost of these operations is estimated between 60-70 million dollars annually. The weight of a neutron source is approximately 6 grams, and its activity is 30 Ci. Due to the high half-life of Am-241 compared to Pu-238, this isotope is used in radioisotope thermoelectric generators (RTGs) in spacecraft. This isotope can also be used to produce other isotopes such as Cm-242 and Pu-238. Cm-242 is used in RTGs due to its high specific thermal capacity (120 $\mathrm{W.g^{-1}}$), while Pu-238 is used in medical applications, such as long-term power supply for artificial hearts. In 1962, Am-241 was first introduced to the market with a current price of \$ 1500 per gram (Ambrosi et al., 2019; Parrington et al., 1996).

While several kilograms of Am-241 are produced annually, this is the only commercially applicable isotope of americium. Though the exact amount of americium production is not well defined, the inventory of Am-243 has been reported between 10 to 100 grams (Seaborg and Loveland, 1991). Am-241 and Am-243 can be produced through the neutron activation of U-239 or Pu-239 in a high flux nuclear reactor.

The content of americium after irradiation of standard PWR and BWR fuel assemblies is approximately 600 grams (0.09%) and 220 grams (0.07%) respectively. Commercial nuclear reactors produce kilogram quantities of the two americium isotopes, americium-241 and americium-243, with the specific isotope ratios being dependent on the reactor burn-up. The DOE Savannah River reactors in the United States produced approximately 9 kilograms of a Am-243, Cm-244 mixture over a period of 10 years. About 1 kilogram mixture of Am-241 and Am-243 was also recovered during the reprocessing of Shippingport blanket fuel at the Hanford DOE site in the United States. Approximately 30 kilograms of americium remain as waste at the Hanford DOE site, while liquid wastes from the Plutonium Uranium Extraction (Purex) process are also considered as potential sources of americium (Miguirditchian et al., 2020). However, The production of americium in research reactors is typically limited to quantities of only a few grams. Americium is primarily produced in research reactors through the neutron activation of target materials, which often involves the irradiation of uranium or other actinides. The specific amount of Am-241 produced can vary significantly depending on the reactors design and operational parameters. The National Nuclear Laboratory (NNL) in the UK has successfully produced Am-241 at the gram scale and is planning to expand to industrial-scale production. Currently, this facility is the only one in the UK capable of producing Am-241, with plans to significantly increase output by 2027 (`https://www.nnl.co.uk/innovation-science-and-technology/showreel/collaborations/space-exploration`). High-flux reactors, such as the High Flux Isotope Reactor at Oak Ridge and the Heinz Maier-Leibnitz reactor in Germany, are capable of producing significant amounts of various isotopes, including Am-241, due to their high neutron flux capabilities

To date, there has been a lack of research in our country concerning the production of these valuable and widely utilized radioisotopes, despite their significant technological importance in the processes of production, separation, and purification. Reactor power, operating schedule, cooling time, and fuel type are crucial parameters for the production of Am-241. In this study, the main focus is on reactor power as the primary parameter influencing the optimal production of Am-241. We have evaluated the feasibility of producing these elements and their activity from the irradiated fuel of typical 1000 MWe and 360 MWe light water reactors, utilizing the ORIGEN code to optimize their production.

2 Research theories

The research was conducted by taking into account certain approximations and simplifications which should be considered in the interpretation and use of the results obtained. The ORIGEN computational code which has been used for calculations, developed for fuel consumption, decay, and radiation dose rate calculations. Key code outputs include isotopes mass in grams, total radiation and alpha radiation in curies, thermal power in watts, neutron absorption, fission, instantaneous fission, (α, n) reactions, and photon counts in 18 energy groups (Waters, 2002; Bell, 1973; Wilson et al., 1997).

3 Results and Discussion

3.1 *Mass and activity calculations of produced Am-241 in 1000 MWe reactor*

Calculations were carried out for a fuel assembly based on its irradiation history in a typical 1000 MWe reactor and considering irradiation periods of 1091 days and cooling periods of 4, 10, 25, and 50 years. The mass and activity of the produced americium from a fuel assembly with a fuel burnup of 43000 megawatt-days per ton in a typical 1000 MWe reactor are presented in Figs. 1 and 2.

3.2 *Calculations of mass and activity of Am-241 produced in a typical 360 MWe reactor assuming 50% fuel consumption*

This reactor has a thermal power of 1130 megawatts, which produces this power from ceramic uranium with three different enrichments: 2.44%, 2.78% and 3.48%. The fuel characteristics of this reactor are considered similar to 1000 MWe reactor. The weight percentage of uranium isotopes in the fuel cannot be more than 88%. According to this article, some of the most important specifications of this reactor are listed in Table 1.

Table 1: Specifications of a typical 360 MWe reactor.

Parameter	Value
Components of a fuel assembly with an average enrichment of 3% (kg)	^{235}U (9.91)/ ^{238}U (320.33)/ O (44.61)
Weight of a standard fuel assembly (kg)	374.8
The number of fuel assemblies	121
The total weight of Uranium 235 in the core (kg)	1199
Total core weight (kg)	45351
The duration of reactor operation in one cycle	354 days

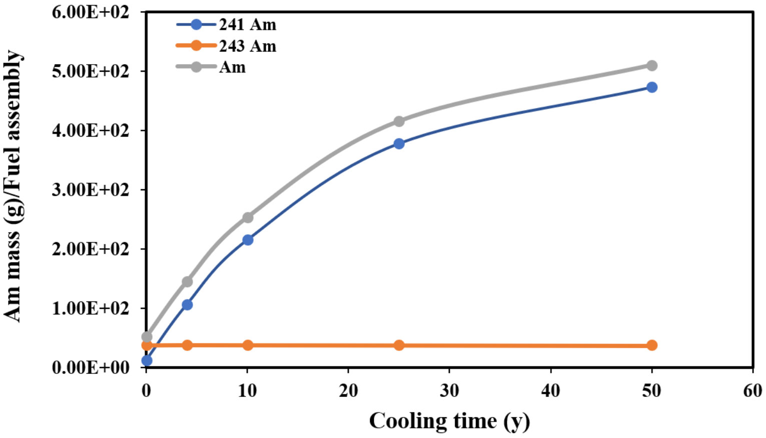

Figure 1: The quantity of Americium production for one standard fuel assembly (g).

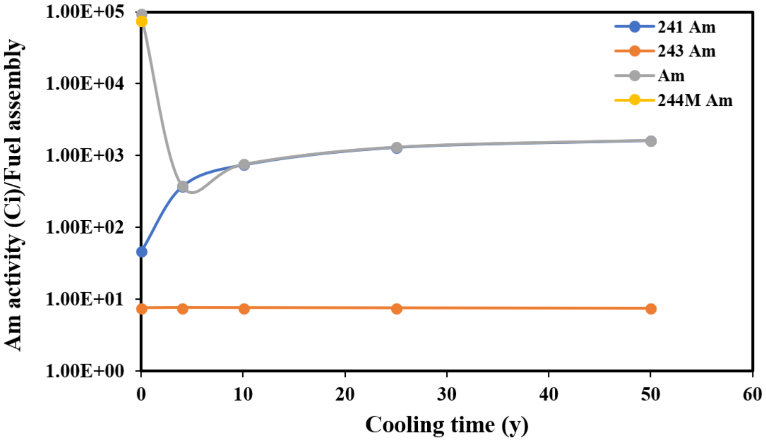

Figure 2: The activity of Americium isotopes for one standard fuel assembly (Ci).

For a fuel assembly operating during 354 days at a power of 1130 megawatts, the amount of 4066 grams of uranium-235 is consumed, which is equivalent to 41.04% of its fuel consumption. In the process of operation during 510 days, the fuel consumption will reach 59.13%, which is the maximum fuel consumption of the assembly. Calculations for the operation during 510 days, has also been done. The mass and activity of amersium produced for one fuel assembly out of 121 core fuel assemblies are shown in Tables 2 and 3, respectively.

3.3 Comparison of Am-241 Production per Unit Mass

In this section, a comparison is made of the production and activity of americium per kilogram of spent uranium in 1000 MWe and 360 MWe reactors. The results are presented in Figs. 3 and 4. As seen, the amount of americium production in the 1000 MWe reactor is higher, as expected, compared to the 360 MWe reactor.

In another work, published by the authors (Boustani et al., 2023), the feasibility study of producing this nuclide using the irradiated fuels of a typical research reactor for two powers of 5 and 10 megawatts and the target of uranium with 99.7% by weight of U-238, which exists at the end of the enrichment cycle has been done using the ORIGEN code. The results of those calculations show that the production efficiency of trans-uranium radionuclide in 10 MWe power is many times higher than 5 MWe. Also, the efficiency of trans-uranium radionuclide production using the target irradiation of depleted uranium is about 2 times higher than the radionuclide produced form the irradiated

Table 2: The quantity of Am production for one standard fuel assembly (g).

	Cooling (yr)				
Radionuclide	0	4	10	25	50
Am-241	2.69E+00	2.99E+01	6.09E+01	1.07E+02	1.34E+02
Am-243	1.60E+00	1.60E+00	1.60E+00	1.60E+00	1.59E+00
Am	4.34E+00	3.16E+01	6.26E+01	1.09E+02	1.36E+02

Table 3: The activity of Am isotopes for one standard fuel assembly (Ci).

	Cooling (yr)				
Radionuclide	0	4	10	25	50
Am-241	9.23E+00	1.03E+02	2.09E+02	3.68E+02	4.60E+02
Am-243	3.18E-01	3.19E-01	3.19E-01	3.19E-01	3.18E-01
Am	6.27E+03	1.04E+02	2.10E+02	3.69E+02	4.61E+02

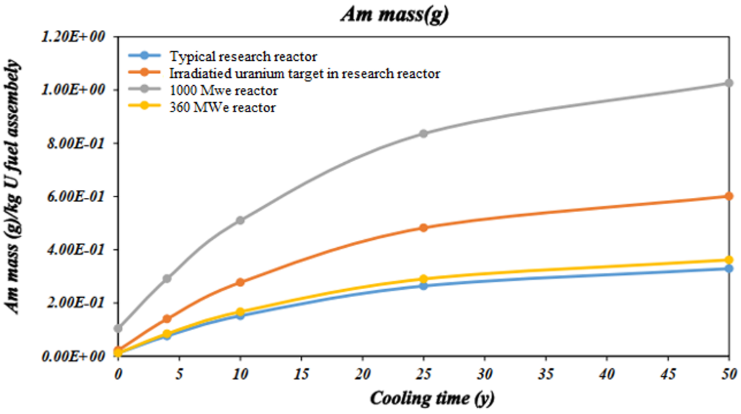

Figure 3: Am production.

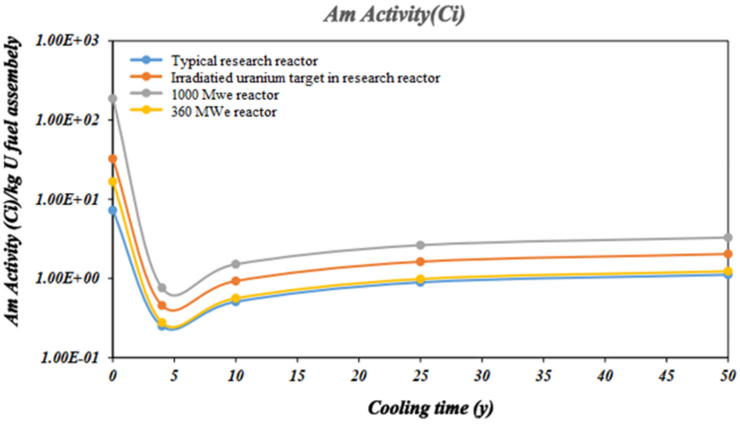

Figure 4: Am activity.

fuel of the typical 5 MWe research reactor. By comparison of those results with this research, it's concluded that the amount of Am-241 production in a 1000 MWe power reactor is about 1.71-4.64 times higher than the target irradiation in the research reactor.

4 Conclusions

In the present work, calculations related to the production and activity of the radioisotope americium-241 from irradiated fuel in the 1000 MWe and 360 MWe reactors have been conducted and the results have been compared. The results indicate that the amount of americium increases with cooling time. Additionally, the production of Am-241 per one kg of consumed uranium in the 1000 MWe reactors is approximately 2.85-8.97 times higher than that of the 360 MWe, and 1.71-4.64 times higher than the target irradiation in the typical research reactor, depending on the cooling period. Regarding the activity, the corresponding values range from 11.2-2.67 and from 1.61-5.75, respectively. Hence, irradiated fuel from the 1000 MWe reactor can be a suitable source for the extraction of this isotope.

Conflict of Interest

The authors declare no potential conflict of interest regarding the publication of this work.

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