

# Impact of Americium-241 (n, $\gamma$ ) Branching Ratio on SFR Core Reactivity and Spent Fuel Characteristics

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## Impact of Americium-241 ( $n,\gamma$ ) Branching Ratio on SFR Core Reactivity and Spent Fuel Characteristics

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### INTRODUCTION

An accurate prediction of core physics and fuel cycle parameters largely depends on the order of details and accuracy in nuclear data taken into account for actual calculations.  $^{241}\text{Am}$  is a major gateway nuclide for most of the minor actinides and thus an important nuclide for core physics and fuel-cycle calculations. The  $^{241}\text{Am}(n,\gamma)$  branching ratio (BR) is in fact energy dependent (see Fig. 1), therefore, it is necessary to take into account the spectrum effect on the calculation of the average BR for the full-core depletion calculations. Moreover, the accuracy of the BR used in the depletion calculations could significantly influence the core physics performance and post irradiated fuel compositions [1].

The BR of  $^{241}\text{Am}(n,\gamma)$  in the ENDF/B-VII.0 library is relatively small and flat in the thermal energy range, gradually increases within the intermediate energy range, and even becomes larger in the fast energy range. This indicates that the properly collapsed BR for fast reactors could be significantly different from that of thermal reactors. The evaluated BRs also differ from one evaluation to another. As seen in Table I, average BRs for several evaluated libraries calculated by means of a fast spectrum are similar but have some differences. While some of widely available and used depletion codes, such as ORIGEN in SCALE [6], have a capability to calculate a variable BR based on the system neutron spectrum, most of the currently available depletion codes still use a pre-determined single value BR for each library. However, ideally it should be determined on an on-the-fly basis like that of one-group cross sections. These issues provide a strong incentive to investigate the effect of different  $^{241}\text{Am}(n,\gamma)$  BRs on core and spent fuel parameters.

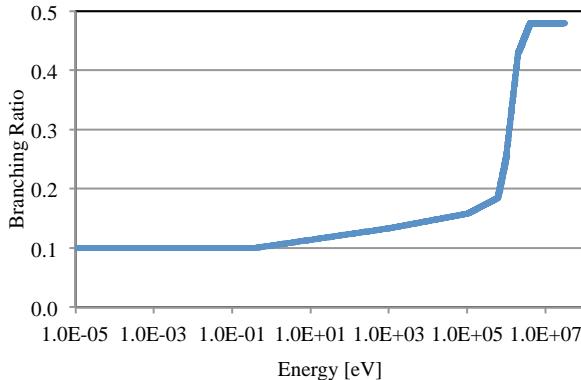


Fig. 1. Energy dependent ENDF/B-VII.0  $^{241}\text{Am}(n,\gamma)$  branching ratio (BR) to  $^{242m}\text{Am}$ .

Table I. Calculated average  $^{241}\text{Am}(n,\gamma)$  branching ratios (BR) to  $^{242m}\text{Am}$  using several evaluated libraries by weighting energy dependent  $^{241}\text{Am}(n,\gamma)$  reaction rates in a SFR core.<sup>a</sup>

Type of Evaluation	BR
ENDF/B-VII.0	0.1581
ENDF/B-VII.1	0.1581
JEFF-3.2	0.1675
JENDL-4.0	0.1519
TENDL	0.1559
CENDL-3.1	0.2654

This paper investigates the impact of the  $^{241}\text{Am}(n,\gamma)$  BR on the results of sodium-cooled fast reactor (SFR) full-core based fuel-cycle calculations. The analysis is performed by gradually increasing the value of BR from 0.15 to 0.25 and studying its impact on the core reactivity and characteristics of SFR spent fuels over extended storage times (~10,000 years).

### MODELING AND PROBLEM DESCRIPTIONS

The SFR fuel-cycle analyses coupled with whole core reactor physics calculations are performed with the REBUS-3 code [2]. The hexagonal-z geometry option is used for modeling a 3D SFR core by applying the periodic boundary condition in the radial plane in order to represent a one third core rotational symmetry. Fig. 2 shows the radial layout of the full-core model. The radial core configuration is based on the S-PRISM design with metallic driver and blanket fuels [3], however, the inner blankets are replaced with driver assemblies in order to reduce the enrichment of the fuel. Neither driver nor blanket assembly shuffling is considered in the model. This SFR is considered to produce 1000 MWth and has thermal efficiency of 40%, cycle length of 530 EFPD, and average discharge burnup of ~49 GWd/t. The residence time of driver assemblies is set to 3 cycles ( $3 \times 530$  EFPD) whereas that of outer blanket assemblies is set to 4 cycles. The radial enrichment variation is assigned by subdividing the radial driver region into three zones, i.e., inner ( $1^{\text{st}} - 4^{\text{th}}$  rows), middle ( $5^{\text{th}} - 7^{\text{th}}$  rows), and outer ( $8^{\text{th}}$  and  $9^{\text{th}}$  rows) zones, in order to flatten the core power distribution. A set of multigroup (33 groups) microscopic cross sections for the nodal diffusion calculations is obtained with the MC<sup>2</sup>-3 code [4] using ENDF/B-VII.0 data.

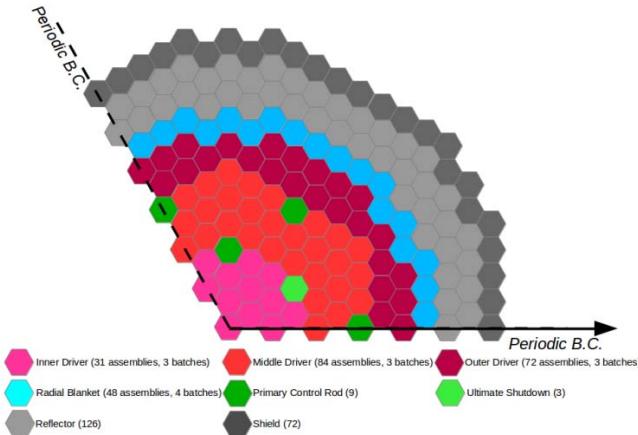


Fig. 2. SFR core model radial layout.

The analysis is performed by gradually increasing the value of BR from 0.15 to 0.25 and studying its impact on the core reactivity and characteristics of SFR spent fuels over extended storage times (~10,000 years). ORIGEN-2.2 [5] is used for estimating their activity, decay heat and radiotoxicities. Both uranium/plutonium (U/Pu) and uranium/transuranic (U/TRU) fuels are examined utilizing the feed composition given in Table II, which corresponds to the discharged TRU composition from the PWR 4.2 % UOX assembly irradiated over 50 GWe·t (cooled for 5 years) calculated with the TRITON module in SCALE 6.1 [6]. (Note that only the Pu composition in Table II is used for U/Pu fuel.) The equilibrium cycle condition is searched by adjusting Pu/TRU feed enrichment so that the core can maintain critical throughout the specified cycle length.

Table II. TRU feed composition used for calculations.

TRU Composition [wt%]	
<sup>237</sup> Np	5.206
<sup>238</sup> Pu	2.349
<sup>239</sup> Pu	47.650
<sup>240</sup> Pu	21.840
<sup>241</sup> Pu	10.660
<sup>242</sup> Pu	6.521
<sup>241</sup> Am	3.398
<sup>242m</sup> Am	0.006
<sup>243</sup> Am	1.715
<sup>242</sup> Cm	9.779E-05
<sup>243</sup> Cm	0.005
<sup>244</sup> Cm	0.594
<sup>245</sup> Cm	0.054
<sup>246</sup> Cm	0.007

## RESULTS

The analysis is carried out by performing full core SFR fuel-cycle calculations in the equilibrium mode by increasing the BR from 0.15 to 0.25 with the step size of

0.025. Note that in this discussion the BR means the BR to <sup>242m</sup>Am. The other counterpart, i.e., (1-BR) which goes to ground state <sup>242</sup>Am, also changes simultaneously.

Table III presents reactivity swing and average feed enrichment of both U/Pu and U/TRU SFR cores with different values of the BR. As seen in this table, as BR increases, the reactivity swing and fuel enrichment decreases. Such a trend is primarily caused by the increase of <sup>242m</sup>Am with increasing BR. Since <sup>242m</sup>Am is the fissile nuclide having a very high fission cross section, once it is produced by a neutron capture, it acts as an excellent source of the fission comparable to <sup>239</sup>Pu. The differences of reactivity swing and feed enrichment relative to the change in BR are more emphasized in U/TRU fuel core because of its larger <sup>241</sup>Am content in the fuel than the U/Pu core.

Table III. Reactivity swing and average feed enrichment with different BRs.

Fuel Type	U/Pu		U/TRU	
	BR	$-\Delta k/k'$ [%]	Enrichment [wt%]	$-\Delta k/k'$ [%]
0.15	1.588	16.226	1.035	17.943
0.175	1.581	16.221	1.017	17.929
0.2	1.571	16.211	0.997	17.913
0.225	1.563	16.206	0.977	17.898
0.25	1.555	16.201	0.959	17.883

Tables IV and V show the relative difference (in percentage) of spent fuel characteristic quantities (activity, decay heat, ingestion and inhalation radiotoxicities) from those obtained with BR=0.15 at different storage times. The negative sign of the relative difference indicates the characteristic quantity is smaller than that of BR=0.15. As seen in these tables, most of characteristic quantities for BR greater than and equal to 0.175 are smaller than those of BR=0.15. This difference is mostly caused by <sup>242</sup>Cm, which is highly radioactive, and one of the key contributors to the total decay heat and radiotoxicities within heavy metals. The smaller BR leads to a larger ground state <sup>242</sup>Am production rate during irradiation, which has a very short half-life (~16 hours) and decays (beta decay) to <sup>242</sup>Cm (half-life = ~163 days) with a high probability (~ 83%). One can also recognize that the total activity for larger BRs at 100 years is slightly larger compared to the one for BR=0.15. The larger BR, on the other hand, results in an increase in the production of <sup>242m</sup>Am, which has a relatively long half-life (141 years). However, it eventually decays to <sup>242</sup>Am, which further decays to <sup>242</sup>Cm. The sum of activities for <sup>242m</sup>Am, <sup>242</sup>Am, and <sup>242</sup>Cm at 100 years accounts for more than 1% of the total activity in the U/Pu case and 3% in the U/TRU case. Thus, it can be implied larger BRs lead to a delayed impact on the total activity. The differences of characteristic quantities from those for the smallest BR become negligibly small after 10,000 years.

## CONCLUDING REMARKS

This paper analyzed the impact of the  $^{241}\text{Am}(n,\gamma)$  BR on the core reactivity and spent fuel characteristics of SFR full-core based fuel-cycle calculations. Since the  $^{241}\text{Am}(n,\gamma)$  BR is energy dependent, the average BR value for the depletion calculations is highly problem-dependent. Therefore, the impact of the BR variation was investigated. Overall the effect of the BR variation (0.15 – 0.25) on those parameters was small; however it was not negligibly small in particular for U/TRU fuel due to its  $^{241}\text{Am}$  content. The reactivity swing and feed enrichment decreased with increasing BR due to the increase in the  $^{242m}\text{Am}$  production which possesses very good fissile properties. The impact of the BR variation within 100 years after the irradiation was primarily characterized by activities of  $^{242}\text{Cm}$ ,  $^{242}\text{Am}$  and  $^{242m}\text{Am}$ .

For the precise estimation of core reactivity and spent fuel characteristics, it is necessary to perform the same type of calculations with each evaluated library, analyze how accurately each library is expected to produce the results, and if necessary, re-evaluate the BR values. Also the estimation of the average BR may need to be performed in every burnup state point, like the generation of one-group cross sections, in order to take into account possible spectrum change due to the transmutation.

## ENDNOTES

<sup>a</sup>  $\sigma_{\gamma}^g$  and  $(\text{BR}^g \cdot \sigma_{\gamma}^g)$  in 33-group structure were calculated with NJOY99.0 [7]. Then the average BR was calculated by weighting 33-group energy spectrum obtained from REBUS-3 calculations.

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Table IV. Relative difference of spent fuel characteristic quantities in percentage from those obtained with BR=0.15 (U/Pu fuel).

BR	Type of Quantity	0 yr	5 yr	10 yr	100 yr	10,000 yr
0.175	Activity	0.001	-0.004	-0.001	0.127	0.000
	Decay Heat	-0.010	-0.090	-0.107	0.001	-0.007
	Ingestion Radiotoxicity	-0.108	-0.150	-0.144	-0.016	0.000
	Inhalation Radiotoxicity	-0.429	-0.193	-0.177	-0.018	0.000
0.2	Activity	0.005	-0.023	-0.017	0.243	-0.038
	Decay Heat	-0.018	-0.198	-0.235	-0.025	-0.040
	Ingestion Radiotoxicity	-0.209	-0.309	-0.304	-0.054	-0.040
	Inhalation Radiotoxicity	-0.912	-0.400	-0.375	-0.059	-0.039
0.225	Activity	0.010	-0.022	-0.020	0.372	-0.038
	Decay Heat	-0.022	-0.288	-0.337	-0.024	-0.054
	Ingestion Radiotoxicity	-0.316	-0.454	-0.459	-0.067	-0.040
	Inhalation Radiotoxicity	-1.364	-0.590	-0.566	-0.074	-0.039
0.25	Activity	0.016	-0.027	-0.020	0.371	-0.038
	Decay Heat	-0.017	-0.288	-0.342	-0.024	-0.047
	Ingestion Radiotoxicity	-0.310	-0.456	-0.459	-0.068	-0.040
	Inhalation Radiotoxicity	-1.364	-0.590	-0.566	-0.074	-0.039

Table V. Relative difference of spent fuel characteristic quantities in percentage from those obtained with BR=0.15 (U/TRU fuel).

BR	Type of Quantity	0 yr	5 yr	10 yr	100 yr	10,000 yr
0.175	Activity	0.013	-0.012	-0.005	0.246	-0.042
	Decay Heat	-0.014	-0.157	-0.188	-0.036	-0.048
	Ingestion Radiotoxicity	-0.215	-0.250	-0.250	-0.063	-0.043
	Inhalation Radiotoxicity	-0.551	-0.289	-0.281	-0.067	-0.042
0.2	Activity	0.014	-0.037	-0.022	0.489	-0.085
	Decay Heat	-0.041	-0.329	-0.358	-0.077	-0.090
	Ingestion Radiotoxicity	-0.446	-0.502	-0.503	-0.124	-0.085
	Inhalation Radiotoxicity	-1.114	-0.577	-0.564	-0.132	-0.084
0.225	Activity	0.032	-0.057	-0.039	0.725	-0.127
	Decay Heat	-0.051	-0.504	-0.549	-0.114	-0.131
	Ingestion Radiotoxicity	-0.652	-0.754	-0.750	-0.190	-0.127
	Inhalation Radiotoxicity	-1.667	-0.863	-0.841	-0.202	-0.126
0.25	Activity	0.039	-0.070	-0.056	0.976	-0.167
	Decay Heat	-0.069	-0.677	-0.737	-0.155	-0.180
	Ingestion Radiotoxicity	-0.875	-0.997	-0.999	-0.252	-0.168
	Inhalation Radiotoxicity	-2.224	-1.148	-1.121	-0.268	-0.165