The study of U-232 accumulation in reprocessed uranium for fast reactor fuel cycle

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Abstract.

One of the main objects of fast reactor nuclear fuel cycle radiation safety is fuel assembly handling. In case of closed nuclear fuel cycle fresh fuel assemblies will be produced from regenerated uranium and plutonium.

Uranium-232 is produced and accumulated in fuel assemblies during the irradiation. One of the U-232 decay products is Tl-208 which emits high energy gamma radiation. In addition, uranium-232 can't be chemically separated from reprocessed uranium. Thereby, the uranium-232 content in reprocessed fuel is very important for fuel cycle radiation safety.

The main ways of uranium-232 production are (n,2n) and (n,3n) reactions on several nuclides. Their contribution to U-232 production depends on their initial content in the fuel. These reactions have neutron energy threshold about 1 MeV.

The difficulty of calculating uranium-232 accumulation is caused by threshold reactions cross sections uncertainties. The evaluation of these cross sections in different libraries can vary by an order or even more.

The paper presents the results of the study into the effect of reaction cross section uncertainties in some modern nuclear data libraries on uranium-232 content and dose rate for reprocessed uranium in fuel assemblies. Fuel cycle scenarios with different fuel compositions, irradiated fuel cooling and fresh fuel storage before irradiation time are considered.

Key Words: closed nuclear fuel cycle, reprocessed uranium, radiation safety, nuclear reaction chains.

1. Introduction

One of the impediment to use the reprocessed uranium again in closed nuclear fuel cycle is the production of ^{232}U. Therefore, it is important to be able to calculate the radiation characteristics of reprocessed uranium with ^{232}U. But there are some cross section uncertainties in the value of (n,2n) and (n,3n) reactions.

This paper provides an analysis of effect of these uncertainties to calculation of 232U production in the fast reactor.

2. Baseline information

There was the test model of sodium fast reactor used for analysis of contribution of different reaction chains to ^{232}U production. To analyze effect of initial fuel condition to ^{232}U production, two different reactor zones considered:

- Bottom axial blanket with depleted uranium oxide fuel;
- Core assembly with MOX fuel.

Initial isotopic composition for two zones considered provided in table 1.

TABLE 1 – INITIAL ISOTOPIC COMPOSITION OF FUEL IN ZONES, %

Nuclide	Axial blanket	Core
^{235}U ^{238}U	0.1% 99.9%	0.1% 99.9%
ΣU	100.0%	78.8%
^{238}Pu ^{239}Pu ^{240}Pu ^{241}Pu ^{242}Pu ^{241}Am		1.5% 61.5% 24.8% 7.2% 5.0% 0.4%
ΣPu		21.1%

Reactor campaign carried in this work is four irradiation intervals, 330 days each with refuelling intervals 35 days.

The main reaction chains tending to ^{232}U production are listed below:

1. $^{238}U \xrightarrow{n,2n} {}^{237}U \xrightarrow{\beta-} {}^{237}Np \xrightarrow{n,2n'} {}^{236m}Np \xrightarrow{\beta-} {}^{236}Pu \xrightarrow{\alpha} {}^{232}U$
2. $^{238}Pu \xrightarrow{\alpha} {}^{234}U \xrightarrow{n,3n} {}^{232}U$
3. $^{238}Pu \xrightarrow{n,3n} {}^{236}Pu \xrightarrow{\alpha} {}^{232}U$
4. $^{237}Np \xrightarrow{n,2n'} {}^{236m}Np \xrightarrow{\beta-} {}^{236}Pu \xrightarrow{\alpha} {}^{232}U$
5. $^{236}Pu \xrightarrow{\alpha} {}^{232}U$
6. $^{234}U \xrightarrow{n,3n} {}^{232}U$
7. $^{235}U \xrightarrow{\alpha} {}^{231}Th \xrightarrow{\beta-} {}^{231}Pa \xrightarrow{n,\gamma} {}^{232}Pa \xrightarrow{\beta-} {}^{232}U$
8. $^{231}Pa \xrightarrow{n,\gamma} {}^{232}Pa \xrightarrow{\beta-} {}^{232}U$

It is noted that the major reactions in the reaction chains is radioactive decay and (n,2n) and (n,3n) reactions. Cross sections of these reactions have quite significant uncertainties, due to they have high an energy reaction threshold, i.e. for $^{238}Pu(n,3n)^{236}Pu$ it is about 13.5 MeV.

From the other hand, radioactive decay data and neutron capture cross-sections were accepted the same for all libraries, to highlight the impact of (n,2n) and (n,3n) reactions uncertainties to calculation results.

For the analysis of uncertainties impact to calculation of ^{232}U production, the following libraries have been considered: ABBN-93[1], ABBN-RF[2], EAF-97[3] и EAF-2010[4]. Assessment of chains contribution to the production of ^{232}U were determined by the computer code SKIF based on certified and verified isotopic kinetics calculation program CARE[5].

3. U-232 accumulation in uranium axial blanket

Assessment of ^{232}U accumulation in the fast reactor uranium blanket, made with various libraries, provided in Table 2. The units are percent ^{232}U in whole U mass.

TABLE 2 – ASSESSMENT OF ^{232}U ACCUMULATION IN BLANKET, %

Time	1 mc[1]		2 mc		3 mc		4 mc
Library	330[2]	35	330	35	330	35	330
ABBN-93	2.61E-11	3.28E-11	1.89E-10	2.16E-10	6.03E-10	6.64E-10	1.36E-09
ABBN-RF	1.26E-10	1.66E-10	1.02E-09	1.19E-09	3.40E-09	3.76E-09	7.82E-09
EAF-97	9.79E-11	1.28E-10	7.87E-10	9.12E-10	2.61E-09	2.89E-09	6.01E-09
EAF-2010	2.82E-11	3.55E-11	2.06E-10	2.36E-10	6.62E-10	7.29E-10	1.50E-09

From the data in Table 2, the following can be concluded: the difference in ^{232}U content assessment is up to 6 times, while the two groups of similar estimates can be distinguished – ABBN-93/EAF-2010 with a discrepancy of about 10% and ABBN-RF/EAF-97 with a discrepancy of about 30%.

To identify the causes of these differences let us compare contribution of several chains to ^{232}U production during irradiation and decay. This comparison is shown in Table 3.

TABLE 3 – CONTRIBUTION OF CHAINS TO ^{232}U PRODUCTION, %

Chain		Time	1 mc		2 mc		3 mc		4 mc
		Library	330	35	330	35	330	35	330
1	$^{238}U \xrightarrow{n,2} {}^{237}U \xrightarrow{\beta-} {}^{237}Np \xrightarrow{n,2n} {}^{236m}Np \xrightarrow{\beta-} {}^{236}Pu \xrightarrow{\alpha} {}^{232}U$	ABBN-93	75.8	0	15.3	0	6.898	0	4.1
		ABBN-RF	94.2	0	16.8	0	7.275	0	4.3
		EAF-97	92.6	0	16.7	0	7.226	0	4.2
		EAF-2010	77.7	0	15.5	0	6.946	0	4.1
7	$^{235}U \xrightarrow{\alpha} {}^{231}Th \xrightarrow{\beta-} {}^{231}Pa \xrightarrow{n,\gamma} {}^{232}Pa \xrightarrow{\beta-} {}^{232}U$	ABBN-93	23.2	0	3.25	0	1.103	0	0.52
		ABBN-RF	4.9	0	0.59	0	0.194	0	0.09
		EAF-97	6.2	0	0.77	0	0.252	0	0.12
		EAF-2010	21.5	0	2.98	0	1.005	0	0.47
5	$^{236}Pu \xrightarrow{\alpha} {}^{232}U$	ABBN-93	0	98.1	31.2	99.1	50.2	99.4	60.1
		ABBN-RF	0	98.9	34.1	99.4	52.8	99.6	62.2
		EAF-97	0	98.9	33.9	99.4	52.5	99.6	61.9
		EAF-2010	0	98.2	31.5	99.1	50.5	99.4	60.4
4	$^{237}Np \xrightarrow{n,2n'} {}^{236m}Np \xrightarrow{\beta-} {}^{236}Pu \xrightarrow{\alpha} {}^{232}U$	ABBN-93	0	0	42.4	0	36.6	0	31.5
		ABBN-RF	0	0	46.4	0	38.5	0	32.6
		EAF-97	0	0	46.1	0	38.3	0	34.5
		EAF-2010	0	0	42.8	0	36.8	0	31.7
8	$^{231}Pa \xrightarrow{n,\gamma} {}^{232}Pa \xrightarrow{\beta-} {}^{232}U$	ABBN-93	0	0	7.05	0	4.7	0	3.3
		ABBN-RF	0	0	1.29	0	0.83	0	0.56
		EAF-97	0	0	1.67	0	1.07	0	0.73
		EAF-2010	0	0	6.45	0	4.28	0	2.97

[1] here and later: micro-campaign, irradiation interval

[2] the duration of irradiation or decay interval, days

It can be mentioned, that the chains, tending to ^{232}U production during first irradiation interval is chains 1 and 7, due to concentration of initial isotopes of other chains is zero at the time of first irradiation start. Also must be mentioned, that the absolute value of chain 7 is equivalent between all cases, because decay and capture speed is identical for all libraries in this study.

The differences of relative contribution of this chain depends on contribution of chain 1. There are reactions (n,2n) on ^{238}U and ^{237}Np nuclei in this chain. The cross sections of these reactions provided in Table 4.

TABLE 4 – CROSS CESTIONS OF (N,2N) REACTIONS ON ^{238}U AND ^{237}NP, BARN

Library	^{238}U (n,2n) ^{237}U	^{237}Np (n,2n') ^{236m}Np
ABBN-93	6.93E-04	2.61E-05
ABBN-RF	7.50E-04	1.45E-04
EAF-97	6.87E-04	1.21E-04
EAF-2010	7.09E-04	2.82E-05

As it is shown in Table 4, differences in ^{238}U (n,2n) ^{237}U cross section between this libraries is below 10%, and it can't be cause huge difference in ^{232}U concentration calculation. Cross section of (n,2n) reaction on ^{237}Np difference is up to 6 times, while the two groups of similar estimates can be distinguished – ABBN-93/EAF-2010 with a discrepancy of about 10% and ABBN-RF/EAF-97 with a discrepancy of about 20%.

With the accumulation of neptunium while irradiation contribution of chains 4 and 5 is increased, exceeding 90% by the end of irradiation. This explains the fact that the content of ^{232}U and the cross section of the (n,2n) reaction on ^{237}Np occur the identical patterns. The coefficient of linear correlation between the values of the content of ^{232}U and cross section of the (n, 2n) on ^{237}Np is 0.99753. Thus, the accumulation of ^{232}U determined by neptunium from the second irradiation interval.

The differences in the estimates of the cross section of the reaction (n, 2n) on 237Np caused by the fact that this reaction is branching:

- With the probability of 21% product is ^{236}Np with the half-life 115000 years, that decays to ^{236}U with probability 91% and ^{236}Pu with probability 8.9%;
- With the probability of 79% product is ^{236m}Np with the half-life 22.5 hours, that decays to ^{236}U with probability 52% and ^{236}Pu with probability 48%.

The differences of this reaction estimates about 6 times leads to the conclusion that the value of this reaction in various libraries may be stored as a total (n,2n) cross section or in separate cross section for ground and metastable branch of ^{236}Np. The analysis of library files shown that in ABBN-93 and EAF-2010 there are additional separate cross sections of metastable branch. Taking into account this data gives an assessment of total (n,2n) cross section in 1.24E-04 barn for ABBN-93 and 1.60E-04 for EAF-2010. Thus maximum difference in this libraries for ^{237}Np(n,2n) reaction cross section is about 32%.

Thus, crucial for the correct assessment of the accumulation of ^{232}U in the fuel during irradiation and exposure is correctly taking into account the total cross section and branching ratios of the reaction (n, 2n) on ^{237}Np.

Also, note that in the software package SKIF that was used to simulate the isotopic kinetics, reaction branching ratios is stored as a separate library. Thus, in this paper, the correct evaluation of the accumulation of ^{232}U performed using libraries ABBN-RF and EAF-97. For ABBN-93 and EAF-2010, the sum of both reaction branches must be used for correct modeling by SKIF.

4. U-232 accumulation in MOX fuel

Assessment of ^{232}U accumulation in the fast reactor uranium MOX fueled core, made with various libraries, provided in Table 5. The units are percent ^{232}U in whole U mass.

TABLE 5 – ASSESSMENT OF ^{232}U ACCUMULATION IN CORE, %

Time	1 mc		2 mc		3 mc		4 mc
Library	330	35	330	35	330	35	330
ABBN-93	1.61E-09	2.03E-09	1.01E-08	1.15E-08	2.76E-08	3.05E-08	5.37E-08
ABBN-RF	7.29E-09	9.73E-09	5.17E-08	5.99E-08	1.47E-07	1.63E-07	2.93E-07
EAF-97	1.41E-08	1.75E-08	6.75E-08	7.63E-08	1.64E-07	1.79E-07	2.98E-07
EAF-2010	1.59E-09	2.09E-09	1.04E-08	1.20E-08	2.88E-08	3.18E-08	5.65E-08

From the data in Table 5, the following can be concluded: the difference in ^{232}U content assessment is up to 6 times, while the two groups of similar estimates can be distinguished – ABBN-93/EAF-2010 with a discrepancy of about 5% and ABBN-RF/EAF-97 with a discrepancy of about 2%.

To identify the causes of these differences let us compare contribution of several chains to ^{232}U production during irradiation and decay. This comparison is shown in Table 6.

TABLE 6 – CONTRIBUTION OF CHAINS TO ^{232}U PRODUCTION, %

Chain		Time	1 mc		2 mc		3 mc		4 mc
		Library	330	35	330	35	330	35	330
1	$^{238}U \xrightarrow{n,2} {}^{237}U \xrightarrow{\beta-} {}^{237}Np \xrightarrow{n,2n} {}^{236m}Np \xrightarrow{\beta-} {}^{236}Pu \xrightarrow{\alpha} {}^{232}U$	ABBN-93	74.0	0	13.3	0	5.8	0	3.4
		ABBN-RF	98.3	0	15.4	0	6.4	0	3.8
		EAF-97	38.9	0	9.6	0	4.7	0	3
		EAF-2010	83.0	0	14.3	0	6.1	0	3.6
2	$^{238}Pu \xrightarrow{\alpha} {}^{234}U \xrightarrow{n,3n} {}^{232}U$	ABBN-93	21.9	0	3.62	0	1.5	0	0.87
		ABBN-RF	0.068	0	0.01	0	0.004	0	0.002
		EAF-97	6.5	0	1.47	0	0.689	0	0.43
		EAF-2010	0.88	0	0.14	0	0.056	0	0.032
5	$^{236}Pu \xrightarrow{\alpha} {}^{232}U$	ABBN-93	0	99.1	33.4	99.5	51.5	99.6	60.3
		ABBN-RF	0	99.1	37.8	99.5	56.5	99.6	65.3
		EAF-97	0	99.5	43.9	99.7	58.1	99.7	65.3
		EAF-2010	0	99.2	38.9	99.5	56.5	99.6	64.8

TABLE 6 (CONT.) – CONTRIBUTION OF CHAINS TO ^{232}U PRODUCTION, %

4	$^{237}Np \xrightarrow{n,2n'} {}^{236m}Np \xrightarrow{\beta-} {}^{236}Pu \xrightarrow{\alpha} {}^{232}U$	ABBN-93	0	0	39.8	0	33.0	0	27.9
		ABBN-RF	0	0	46.0	0	36.6	0	30.5
		EAF-97	0	0	28.8	0	27.0	0	24.4
		EAF-2010	0	0	42.9	0	34.9	0	29.3
6	$^{234}U \xrightarrow{n,3n} {}^{232}U$	ABBN-93	0	0	8.2	0	6.75	0	5.8
		ABBN-RF	0	0	0.022	0	0.018	0	0.015
		EAF-97	0	0	3.3	0	3.1	0	2.83
		EAF-2010	0	0	0.31	0	0.25	0	0.22
3	$^{238}Pu \xrightarrow{n,3n} {}^{236}Pu \xrightarrow{\alpha} {}^{232}U$	ABBN-93	2.04	0	0.34	0	0.14	0	0.081
		ABBN-RF	0.28	0	0.041	0	0.016	0	0.009
		EAF-97	53.6	0	12.2	0	5.7	0	3.5
		EAF-2010	14.3	0	2.27	0	0.92	0	0.53

In general, for 1st irradiation interval the distribution of the major contributors is like their distribution in case of uranium blanket – one of the main channels is chain 1. The other channels 2 and 3, starting from ^{238}Pu, has significant differences of its contribution – (0.07% - 22%) and (0.3% - 54 %) respectively. These differences are related to the uncertainty of threshold reaction cross sections (n, 3n) in the nuclei ^{234}U and ^{238}Pu. These cross sections are given in Table 7.

TABLE 7 – CROSS CESTIONS OF SOME (N,2N) AND (N,3N) REACTIONS, BARN

Library	^{238}U (n,2n) ^{237}U	^{237}Np (n,2n') ^{236m}Np	^{238}Pu (n,3n) ^{236}Pu	^{234}U (n,3n) ^{232}U
ABBN-93	1.88E-03	7.11E-05	1.05E-08	3.13E-06
ABBN-RF	2.03E-03	3.95E-04	6.61E-09	4.39E-08
EAF-97	1.86E-03	3.29E-04	2.41E-06	8.06E-06
EAF-2010	1.92E-03	7.69E-05	7.27E-08	1.23E-07

As in the previously considered case with uranium blanket, significant differences are observed in the evaluation of (n,2n) cross section on ^{237}Np. The reasons were analyzed and described before, only note that the value of this cross section in the core is increased in comparison with the blanket due to more rigid neutron spectrum in the core.

As for the reactions (n,3n) on the ^{234}U and ^{238}Pu, then their evaluations have considerable differences of up to 2-3 order of value. They are connected with the fact that the estimates of these cross sections performed by the calculations on nuclei models and correspondent experiment were not conducted.

However, as in the case of uranium blanket, by the end of irradiation the major contribution (about 90%) have chains 4 and 5. The coefficient of linear correlation between ^{232}U content and $^{237}Np(n,2n)^{236m}Np$ cross section is 0.98465, which is slightly less than for uranium fuel, since the contribution of the (n,3n) reaction is only 6% even with maximum cross sections evaluations.

Therefor, significant differences in the cross sections (n, 3n) at ^{234}U and ^{238}Pu in pairs ABBN-93 / EAF-2010 and ABBN-RF / EAF-97 does not lead to an increase in discrepancies in the assessment of the amount of uranium-232 above the 2-5% .

5. Conclusion

The presented study shows, that there are some issues in analyzing the ^{232}U content in reprocessed uranium. There are significant differences in the evaluations of some cross sections, but their effect to the calculation result is not trivial.

The first thing to mention is that such calculation must be performed preferably in the steady state, due to effect of ^{236}Pu, ^{238}Pu, ^{237}Np and ^{234}U accumulation to ^{232}U production.

The second is that fact, that even (n,3n) reaction on ^{234}U and ^{236}Pu cross sections have significant differences, their effect to ^{232}U content quite low.

Much more important is to correctly take into account branching reactions, and the method of its storage in libraries, especially while converting libraries from one storage format to another.

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