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CALCULATION OF THE TRANSMUTATION RATES OF Tc-99, I-129 AND Cs-135 IN THE HIGH FLUX REACTOR, IN THE PHÉNIX REACTOR AND IN A LIGHT WATER REACTOR

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ABSTRACT

Transmutation of long-lived fission products is of interest for the reduction of the possible dose to the population resulting from long-term leakage of nuclear waste from waste disposals. Three isotopes are of special interest: Tc-99, I-129 and Cs-135. Therefore, experiments on transmutation of these isotopes in nuclear reactors are planned. In the study described in this report, the possible transmutation rates and mass reductions were determined for experiments in the High Flux Reactor (HFR) located in Petten, the Netherlands, and in Phénix located in France. Also, the rates were determined for a standard Light Water Reactor (LWR). The transmutation rates of these three fission products will be much higher in the HFR than in the Phénix reactor, because both the total flux and the effective cross sections are higher. For thick targets the effective half lives are approximately 3 years, 2 years and 7 years for Tc-99, I-129 and Cs-135 irradiation respectively in the HFR and 22 years, 16 years and 40 years for Tc-99, I-129 and Cs-135 irradiation respectively in the Phénix reactor. The transmutation rates in an LWR are low. Only the relatively large power of an LWR guarantees a large total mass reduction. Especially the transmutation of Cs-135 will be very difficult in the Phénix reactor and LWR clearly shown by the very long effective half lives of 40 and 100 years, respectively.

Contents

1 INTRODUCTION

Transmutation of long-lived fission products is of interest for the reduction of the possible dose to the population resulting from leakage of nuclear waste from waste disposals. Three isotopes are of special interest: Tc-99, I-129 and Cs-135 Therefore, experiments on transmutation of these isotopes in nuclear reactors are planned. In the study described in this report, the possible transmutation rates and mass reductions were determined for experiments in the High Flux Reactor (HFR) located in Petten, the Netherlands, and in Phénix located in France. Also, the rates were determined for a standard LWR. These experiments were modelled by a simple cylindrical geometry. Later studies will determine the rates for real designed experiments This study is only meant to determine which reactor is most suited for such experiments.

2 MATERIALS AND METHOD

2.1 Cross section data

Tc-99 and I-129 data were obtained from the JEF2.2 data file. For I-129, the very recent re-evaluation by ECN was used [1]. For Cs-135, the data from JEF1.1 was used. The data were processed by the NJOY/NSLINK system [2] to the 219 group structure as presented in reference [3] for the HFR and the LWR calculations. For the Phénix calculations, the data were processed in the same way to 26 groups according to the group structure as presented in table 1. This group structure agrees with the 25 group structure used by CEA [4], however, with one additional group for neutron energies above 14.5 MeV and an other group width for the two groups with lowest energies.

group	energy [eV]	Δu
1	$2\cdot 10^7$	0.32
2	$1.45\cdot 10^7$	1.37
3	$3.68\cdot 10^6$	0.5
4	$2.23\cdot 10^6$	0.5
5	$1.35\cdot 10^6$	0.5
6	$8.21\cdot 10^5$	0.5
7	$4.98\cdot 10^5$	0.5
8	$3.02\cdot 10^5$	0.5
9	$1.83\cdot 10^5$	0.5
10	$1.11\cdot 10^5$	0.5
11	$6.74\cdot 10^4$	0.5
12	$4.09\cdot 10^4$	0.5
13	$2.48\cdot 10^4$	0.5
14	$1.50\cdot 10^4$	0.5
15	9120	0.5
16	5530	0.5
17	3360	0.5
18	2040	0.5
19	1230	0.5
20	748	0.5
21	454	0.5
22	275	1.0
23	101	1.5
24	22.6	2.0
25	3.06	2.8
26	0.19	2.0

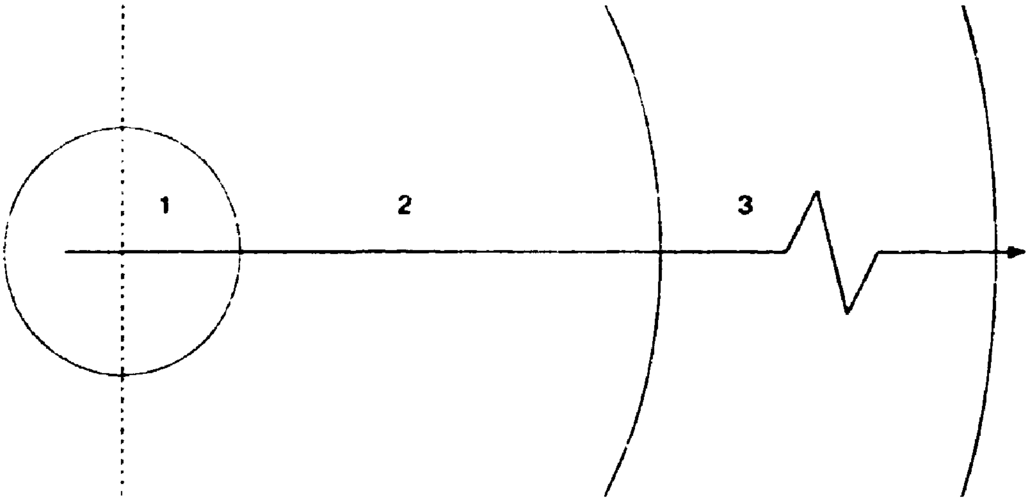
Table 1: Group structure used for the Phénix calculations [4].

2.2 Calculational method

A detailed spectrum in 219 groups was available for the HFR in the core position E5, in which an extremely high thermal flux may be obtained [5]. In figure 1 the configuration for the calculations is shown. The neutron flux per unit of lethargy in the iron center is presented in figure 2, taken from [5].

In the calculations for the experiments in Phénix, we used the flux spectrum in a Neptunium target, surrounded by moderator positioned in the radial blanket of Phénix [6]. The geometrical setup is shown in figure 3. The flux spectrum is presented in figure 4.

The neutron flux per unit of lethargy of an LWR is presented in figure 5 and was calculated for the EPRI benchmark [3].



Material number	Radius [cm]	Description
1	1.0	Iron mockup sample
2	4.46	Moderator
3	25.	Surrounding of HFR core position E5

Figure 1: Experimental setup in the HFR experiments to obtain a high thermal flux [5].

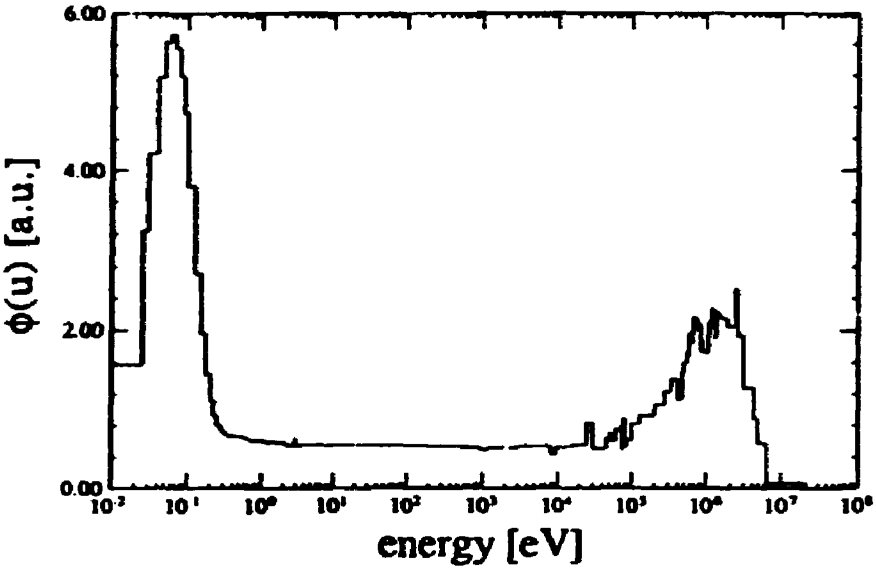


Figure 2: The neutron flux per unit of lethargy as a function of energy in a special geometrical position of the HFR [5].

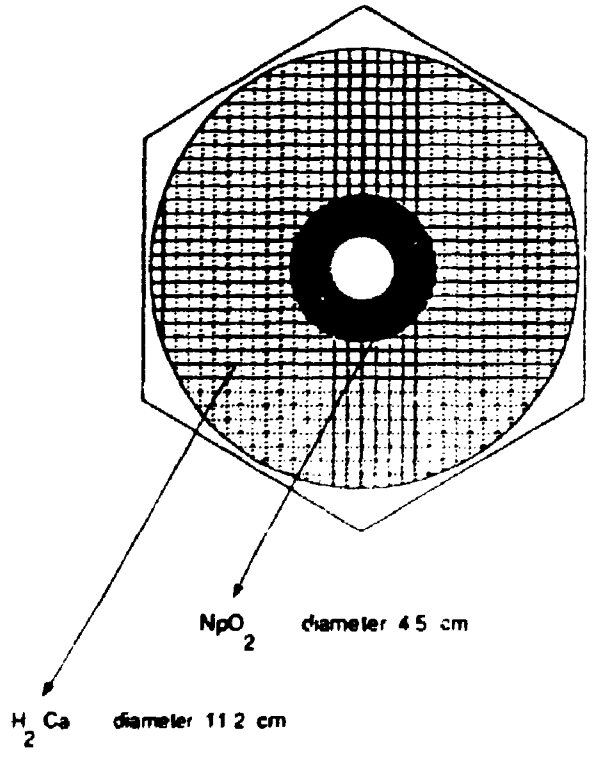


Figure 3: Experimental setup in the Phénix experiments on irradiation of Np 237 as presented in reference [6].

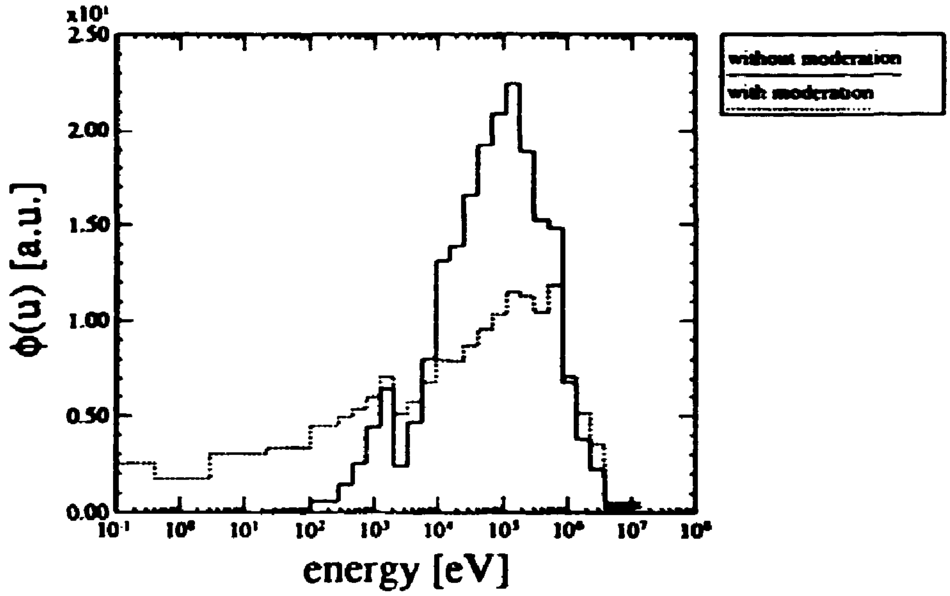


Figure 4: Unmoderated and moderated Phénix neutron flux per unit of lethargy as a function of energy [6].

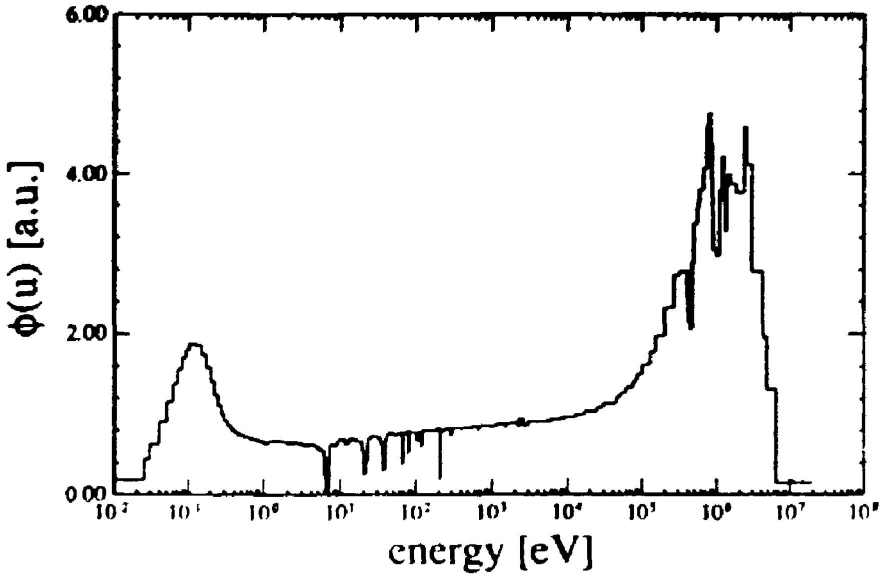


Figure 5: LWR neutron flux per unit of lethargy as a function of energy.

The calculations were made with the PASC-3 system based upon AMPX and SCALE-3 modules [7]. In this system, the Bondarenko method is used for selfshielding calculations in the unresolved range (code BONAMI) and the Nordheim method is used in the resolved region (code NITAWL). The calculations began with a sequence of BONAMI and NITAWL to determine the multigroup selfshielded cross sections in the unresolved and resolved energy regions, respectively. To determine the absorption reaction rate from the 219 or 26 groups cross sections calculated with these codes, we had to collapse with group fluxes which are perturbed by the resonances. But these flux perturbations are not on the output of the codes. In NITAWL however, it is possible to calculate the selfshielding with respect to the unperturbed flux. For BONAMI, the selfshielding with respect to the unperturbed flux cannot be calculated and we have to approximate the selfshielding with respect to the unperturbed flux by the selfshielding with respect to the perturbed flux. This is sufficiently accurate for the energy regions treated by BONAMI. All calculations were made in a simple cylindrical geometry.

With the one-group effective cross section $\sigma(r)$ calculated in this way, several quantities of the transmutation process in cylindrical samples containing Tc-99, I-129 or Cs-135 may be calculated, depending on the radius r of the sample, the total flux ϕ and the total irradiation time t:

- The change in mass equal to $M(t=0)(1 - exp(-\sigma(r)\phi t))$.
- The effective half life equal to $\frac{log(2)}{\phi\sigma(r)}$.

The integrated flux in the HFR is 10^{15} cm^{-2}s^{-1} and the integrated flux in the Phénix experimental setup is approximately $5.3 \cdot 10^{14}$ cm^{-2}s^{-1}. The integrated flux in an LWR is $1.8 \cdot 10^{14}$ cm^{-2}s^{-1}.

The densities of Tc-99 and Cs-135 were taken equal to their pure metal densities which are 11.5 g cm^{-3} and 1.9 g cm^{-3}, respectively, and we assumed that these isotopes are totally separated from other isotopes. It should be noted, however, that the fission product Cesium contains only about 30% Cs-135.

The density of I-129 was taken equal to 2.94 g cm^{-3} assuming Iodide is bound to Sn in SnI_2 with a density of 4.5 g cm^{-3} and Iodide is the fission product Iodide with approximately 80% I-129 and 20% I-127.

3 RESULTS

In tables 2, 3 and 4 the effective cross sections and half lives of Tc-99, I-129 and Cs-135 are presented for the total fluxes as presented in section 2.

radius	effective half life [y]			effective cross section [b]		
[cm]	Tc-99	I-129	Cs-135	Tc-99	I-129	Cs-135
0.00	1.60	1.79	4.85	13.71	12.31	4.53
0.10	2.71	1.81	5.87	8.12	12.16	3.74
0.30	2.91	1.82	6.22	7.56	12.05	3.53
0.70	3.01	1.84	6.42	7.31	11.96	3.42
1.00	3.04	1.84	6.48	7.23	11.93	3.39
2.00	3.10	1.85	6.59	7.10	11.88	3.34
2.25	3.10	1.85	6.61	7.08	11.87	3.33

Table 2: Effective half life and the effective one group cross section as functions of cylinder radius for the HFR with a total flux of 10^{15} $cm^{-2}s^{-1}$.

radius	effective half life [y]			effective cross section [b]		
[cm]	Tc-99	I-129	Cs-135	Tc-99	I-129	Cs-135
0.00	3.73	11.91	13.79	11.12	3.48	3.91
0.10	11.75	12.94	23.54	3.53	3.21	1.76
0.30	15.27	13.88	29.56	2.72	2.99	1.40
0.70	18.32	14.77	34.19	2.26	2.81	1.21
1.00	19.57	15.17	36.00	2.12	2.73	1.15
2.00	21.87	15.93	39.46	1.90	2.60	1.05
2.25	22.25	16.05	39.99	1.86	2.58	1.04

Table 3: Effective half life and the effective one group cross section as functions of cylinder radius for the moderated Phénix spectrum with a total flux of $5.3 \cdot 10^{14}$ $cm^{-2}s^{-1}$.

radius	effective half life [y]			effective cross section [b]		
[cm]	Tc-99	I-129	Cs-135	Tc-99	I-129	Cs-135
0.00	13.25	29.64	44.10	9.22	4.12	2.77
0.10	34.14	31.03	70.54	3.58	3.93	1.73
0.40	40.51	32.11	84.27	3.01	3.80	1.45
0.70	41.96	32.92	93.64	2.72	3.71	1.30
1.00	46.62	33.23	97.07	2.62	3.67	1.26
2.00	49.40	33.74	102.87	2.47	3.62	1.19
2.25	49.82	33.82	103.83	2.45	3.61	1.18

Table 4: Effective half life and the effective one group cross section as functions of cylinder radius for an LWR with a total flux of $1.8 \cdot 10^{14}$ $cm^{-2}s^{-1}$

In tables 5, 6 and 7 the transmuted mass in grams and percentage per cycle of Tc-99, I-129 and Cs-135 in the HFR, the moderated Phénix spectrum and in an LWR are presented assuming a cycle time of 3 years and a pin length of 1 meter.

radius	transmuted mass [g]			transmuted fraction [%]		
[cm]	Tc-99	I-129	Cs-135	Tc-99	I-129	Cs-135
0.00				72.7	68.8	34.9
0.10	19.4	6.31	1.78	53.6	68.4	29.8
0.30	166	56.5	15.3	51.1	68.0	28.4
0.70	884	307	80.9	49.9	67.7	27.7
1.00	1790	625	164	49.5	67.7	27.4
2.00	7070	249	646	48.9	67.5	27.1
2.25	8930	3150	816	48.8	67.5	27.0

Table 5: Transmuted mass in grams and percentage as a function of cylinder radius for the HFR with a total flux of 10^{15} $cm^{-2}s^{-1}$ for a cycle time of 3 years and a pin height of 1 meter.

radius	transmuted mass [g]			transmuted fraction [%]		
[cm]	Tc-99	I-129	Cs-135	Tc-99	I-129	Cs-135
0.00				42.7	16.0	14.0
0.10	5.86	1.37	0.505	16.2	14.9	8.46
0.30	41.4	11.6	3.65	12.7	13.9	6.79
0.70	190	59.4	17.3	10.7	13.1	5.90
1.00	364	118	33.5	10.1	12.8	5.61
2.00	1310	452	123	9.07	12.2	5.13
2.25	1630	568	153	8.92	12.2	5.07

Table 6: Transmuted mass in grams and percentage as a function of cylinder radius for the moderated Phénix spectrum with a total flux of $5.3 \cdot 10^{14}$ $cm^{-2}s^{-1}$ for a cycle time of 3 years and a pin height of 1 meter.

radius	transmuted mass [g]			transmuted fraction [%]		
[cm]	Tc-99	I-129	Cs-135	Tc-99	I-129	Cs-135
0.00				14.5	6.78	4.61
0.10	2.13	0.599	0.173	5.91	6.48	2.90
0.40	28.9	9.27	2.33	5.00	6.27	2.44
0.70	80.0	27.7	6.42	4.52	6.12	2.20
1.00	158	56.0	12.6	4.36	6.07	2.12
2.00	596	221	47.8	4.12	5.98	2.00
2.25	748	279	59.9	4.09	5.96	1.98

Table 7: Transmuted mass in grams and percentage as a function of cylinder radius for an LWR with a total flux of $1.8 \cdot 10^{14}$ cm^{-2}s^{-1} for a cycle time of 3 years and a pin height of 1 meter.

The effective cross sections are much higher for the HFR than for the Phénix and LWR reactors, resulting from the very high thermal flux. So, also the effective half lives in the HFR will be much smaller. The mass fractions transmuted in the HFR for large samples will be approximately 49%, 68% and 27% for Tc-99, I-129 and Cs-135, respectively, when irradiated with a total flux of 10^{15} $cm^{-2}s^{-1}$ for a cycle time of 3 years. The mass fractions transmuted in the Phénix spectrum with moderation will be approximately 9%, 12% and 5% for Tc-99, I-129 and Cs-135, respectively, when irradiated with a total flux of $5.3 \cdot 10^{14}$ $cm^{-2}s^{-1}$ for a cycle time of 3 years.

4 DISCUSSION AND CONCLUSIONS

Several assumptions were made to obtain the one group effective cross sections and the transmutation rates:

1. Only selfshielding of the resonances was calculated, which is not enough because also the flux depression by absorption in the thermal region will be considerable. This flux depression should be calculated as a function of energy with a transport code. The flux depression will be more important for the HFR and the LWR spectrum than for the moderated Phénix spectrum because the flux in the thermal region in the moderated Phénix spectrum is very low. The effective cross sections in the HFR will be lower due to these considerations, but not with a factor three. which is the difference between the effective cross sections in the HFR and in the Phénix reactor.

2. The densities of Tc-99 and Cs-135 were equal to the densities in metal form. which may not be realistic. The selfshielding is nearly proportional to the product of density and radius. So, for other densities the effective cross section may be very easily calculated from the tabulated values (for instance if the density is two times lower, the effective cross section will be equal to the tabulated cross section at one half of the real radius).

3. The influence of other isotopes on the transmutation was not considered. These isotopes may change the flux spectrum or may produce the isotope considered (e.g. Cs-134 may produce Cs-135 when irradiated).

4. In the calculations on the transmuted mass. the density was kept constant. For very high burnup, the densities will be lower and the self shielding effect will be smaller. So. the effective cross sections will increase with burnup.

Although these assumptions should be studied in more detail. one may conclude that the effective cross sections in the HFR shall be higher than in a moderated Phénix spectrum. Because also the total flux in the HFR is higher. the transmutation rate in a HFR will be much higher than in the Phénix reactor. However. one has to be careful to state that the HFR is more suited for transmutation of fission products. because this is also depending on the total amount of available neutrons. which is proportional to the power of the reactor.

Especially the transmutation of Cs-135 will be very difficult in the Phénix reactor and LWR clearly shown by the very long effective half lives of 10 and 100 years. respectively.

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