Neutronic Analyses of Hybrid Systems with TRU Targets

Y. Kadi[1], P.Wydler
Paul Scherrer Institute
CH–5232 Villigen PSI
(Switzerland)

Abstract

For transmutation systems based on externally driven subcritical assemblies with a fast neutron spectrum, there is an incentive to expose the actinides directly to the source neutrons, since these neutrons have higher energies than the fission neutrons. To clarify the influence of the high–energy models on the transmutation effectiveness of such systems, a sensitivity study based on the Phoenix concept, i.e a sodium–cooled system with a minor actinide oxide fuelled target, was performed. The calculations show that the differences arising from the use of different basic data sets are quite as significant as the effects of using different source approximations and hence also deserve attention.

Analyse neutronique de systèmes hybrides d'incinération de cibles de transuranien

Résumé

Pour les systèmes de transmutation à milieu sous–critique rapide et alimentés par une source externe de neutrons, il est avantageux d'exposer directement les actinides mineurs aux neutrons d'évaporation, plus énergétiques que les neutrons de fission. Une étude paramétrique, basée sur le concept Phoenix, a été réalisée afin de déterminer l'influence des modèles de calcul à haute énergie (en particulier le modèle de fission) sur les rendements de transmutation de ces systèmes. Les calculs montrent que les caractéristiques neutroniques du concept Phoenix (spectre neutronique, taux de transmutation,etc.) sont autant sensibles aux incertitudes relatives des données nucléaires de base qu'aux méthodes de calcul à haute énergie.

---

[1] On attachment from CEA, CEN Cadarache, F-13108 St–Paul–lez–Durance

1. General Background

In order to respond to the public concern about nuclear wastes and, in particular, the long–lived high level ones, a French law issued on December 30, 1991 identified the major objectives of research for the next fifteen years in the field of waste management. To comply with the requirements of this law, the CEA (Commissariat à l'Energie Atomique) has launched an important and long term R & D programme, a part of which, called SPIN [1], is devoted to the SeParation and INcineration of these wastes. In a short and mid term perspective, a subprogramme, PURETEX, will aim primarily at reducing the volume of wastes from conventional reprocessing by a factor of 3. In the long term, the subprogramme ACTINEX will be devoted to the separation and transmutation of long–lived elements with the aim of reducing the toxicity of the wastes by a factor of 100 and then 1000 compared to direct disposal.

In the field of transmutation, studies at the CEA are in progress in two directions : validation of the nuclear data which are necessary for incineration studies (cross sections, yields, decay data), parametric studies in view of evaluating the feasibility and the conditions of actinide incineration in conventional power reactors (PWRs or fast reactors). In parallel, the possibilities of hybrid systems, involving a proton accelerator and a subcritical medium, are being investigated in cooperation with the Paul Scherrer Institute (PSI). With its experience in reactor and accelerator–based physics, including the development of the SINQ spallation neutron source, PSI is in a good position to perform physics studies related to such systems.

Since accelerator–based reactor systems are technically more complex and tend to be less economic than normal fission reactors, it is reasonable to design accelerator–based actinide transmutation systems specifically with the object of fissioning the even–neutron minor actinides (^{237}Np, ^{241}Am, etc.) which, due to the threshold in their fission cross section, cannot effectively be transmuted in normal reactors. This thinking leads to accelerator–based transmutation concepts based on fast neutrons. The PSI–CEA activities [2,3] in the field of accelerator–based transmutation are aimed at establishing the scientific basis for assessing the effectiveness of such concepts and resolving related "data and methods" problems.

2. Potential of Fast–Neutron Based Systems

It is obvious that the transmutation effectiveness for actinides is related to the fission–to–capture ratio of the nuclides and, for even–neutron nuclides, therefore significantly increases with neutron energy. Accelerator–based systems offer the possibility, on the one hand, of "hardening" the neutron spectrum beyond the limits of normal fission reactors (possibility of using pure minor actinide fuels, spectrum hardening due to evaporation neutrons which have a higher energy than the fission neutrons) and, on the other hand, of using the high–energy reactions themselves to fission actinides. Concepts with TRU targets incorporate all of these features and therefore appear to be particularly attractive. Examples are the Phoenix concept [4] and the molten salt concept proposed by JAERI [5].

Fission–to–capture ratios for different systems, calculated using a scheme [6] which incorporates the PSI version of the high–energy code HETC and JEF–2.2 data for the neutron transport below 15 MeV, are shown in Table 1. The "D_2O cell" values correspond to a well moderated thermal neutron spectrum, typical for the D_2O moderator of a continous spallation source, and the "FBR" values correspond to a Superphénix type spectrum. Very favourable fission–to–capture ratios are obtained for the Phoenix reference case, and these can be further improved by replacing the reference minor actinide oxide fuel by metal fuel.

Another important aspect is the overall neutron balance of a system. For a closed, long–term system this should be such as to allow the complete conversion of the actinides to fission products. Salvatores et al. [7] have proposed to measure the overall neutron balance in terms of the "fuel neutron production" parameter, here denoted by "p". Unlike other neutron balance parameters, "p" depends on the ratio of neutron induced reactions (fissions, captures, n,2n reactions) to radioactive decays and therefore on the neutron flux.

"p" values for different systems are shown in Table 2. For the "problem nuclides" (e.g. ^{237}Np), the overall neutron balance in an LWR is negative, indicating that the chain of successive transmutations does not provide enough neutrons to support itself. In thermal systems with a very high flux "p" becomes positive, but remains small compared with "p" values in fast systems. Again, the most favourable results are obtained for the Phoenix system.

3. Sensitivity of Basic Parameters to Data and Methods

High–energy nucleon–meson transport codes, such as HETC, have usually been validated with a view to their use in the design of spallation neutron sources for solid–state physics applications [8]. In the context of transmutation, a correct prediction of the neutron source strength in the target is not the only goal. The code has also to be capable of correctly predicting the neutron spectrum, particularly of the evaporation neutrons, and the mass distribution of spallation and fission products, since the individual nuclides are associated with widely differing toxicities and half–lives. One of the models which influences these quantities is the high–energy fission model. Simple code comparisons [9,10] have revealed considerable differences in the total yield and the shape of the mass distribution for both spallation and fission products.

Therefore, to assess the practical importance of nuclear model differencies, a sensitivity study was carried out for the Phoenix reference system, i.e the system with oxide fuel. In particular, calculations were performed for a "subcritical" case, simulating a fission neutron driven subcritical target with the same k_{eff} as that of the reference case, and for an evaporation neutron source driven subcritical target with the high–energy fissions in HETC disabled.

Table 3 gives a comparison, over all energies, of the nuclear interactions taking place in a Phoenix target module with and without high–energy fissions. From this comparison, the following conclusions can be drawn:

1. For the high-energy interactions, there is a 35% reduction in energy deposition when high–energy fission is "switched off". This is due to the lower heat production in the spallation reaction, about 40 MeV per neutron as opposed to typically 100 MeV for fission. However, with 95% of the fissions occuring below 15 MeV (depending on the k_{eff} of the target), an accurate prediction of the fission fragment kinetic energies by the high–energy fission model is not so important in predicting the overall target heating.

2. Neutron production from evaporation (taken to be below 15 MeV) is 3% higher with high–energy fissions disabled. As can be seen in Figure 1, "switching off" high–energy fission results in a noticeable "spectral softening", the mean neutron energy droping by 9% to 3.73 MeV. However, the spectrum remains much harder than a normal fission spectrum.

3. Large differences are also observed for the mass distribution of the high–energy reaction products (see Fig.2) and their toxicity (see Fig.3), indicating a strong sensitivity to the competition, in the de–excitation mode, between evaporation and fission for TRU targets. "Switching off" high–energy fission results in a three to more than ten fold increase in the radiotoxicity of the residual nuclei (the toxicity of the spallation products is greater than that of the fission products). With fission disabled, the toxicity induced by the high–energy reactions, representing only 5% of the total reactions, is of the same order of magnitude as the toxicity induced by the low–energy fissions. Hence, an accurate prediction of the high–energy fissions is essential in estimating the toxicity generated in the target.

Calculations of the total neutron flux spectrum in the target with and without the high–energy fission model were made. The spectra, given in Figure 4, indicate only a small spectral softening when the high–energy fissions are "switched off". A somewhat larger softening of the spectrum is observed, if the spallation source is replaced by a fission source.

Tables 4 and 5 give fission–to–capture ratios and "p" values for different modelling assumptions and basic data sets. It can be seen that the error introduced by the use of a fission neutron source rather than an evaporation neutron source is between 6 – 8% for the "probem nuclei" and that, for neutronic calculations alone, the use of a high–energy fission model is not essential. The differences arising from the use of different basic data sets are quite as significant as the effects of using different source approximations and hence also deserve attention.

Mass changes for the investigated systems are given in Tables 6 and 7. The tabulated values apply for a fresh core (first cycle) and indicate that a high–energy fission model is not needed for burn–up calculations.

Acknowledgments

The analytical work described in this paper is performed in the framework of a doctoral thesis supported by a grant from the CEA and Framatome. It has been carried out at the Laboratory for Reactor Physics and Systems Technology at the Paul Scherrer Institute in collaboration with the CEA.

We gratefully acknowledge the help of Mr. F. Atchison in providing a version of HETC with the high–energy fission model disabled.

References

[1] J-Y Barre, J. Bouchard, "French R & D Strategy for the Back End of the Fuel Cycle",

Proc. of the GLOBAL'93 Int. Conf. and Technology Exhibition on Future Nuclear Systems, Seattle, Washington, U.S.A. (1993)

[2] P. Wydler, R. Chawla, Y. Kadi, "Activities Related to Accelerator–Based Transmutation at PSI", 8th Journées SATURNE on Accelerators Applied to the Nuclear Waste Problem, Saclay, 5–6 May 1994

[3] M. Salvatores et al., "The SPIN Program at CEA: Transmutation Aspects", Proc. of the GLOBAL'93 Int. Conf. and Technology Exhibition on Future Nuclear Systems, Seattle, Washington, U.S.A. (1993)

[4] G.J. van Tuyle et al., "The Phoenix Concept: Proposed Transmutation of Long–Lived Radioactive Wastes to Produce Electric Power", BNL–52279 (1991)

[5] Y. Kato et al., "Accelerator Molten Salt Target System for Transmutation of Long Lived Nuclides", Proc. of the OECD–NEA Specialists' Meeting on Accelerator–Based Transmutation, PSI Würenlingen/Villigen, Switzerland, 24–26 March 1992

[6] S. Pelloni, Y. Kadi, H.U. Wenger, "Present Methods for Physics Calculations of Hybrid Fast Systems at PSI", this meeting

[7] M. Salvatores et al., "A Global Physics Approach to Transmutation of Radioactive Nuclei", Nucl. Sci. Eng., **116**, 1–18 (1994)

[8] F. Atchison, "Data and methods for the design of accelerator–based transmutation systems", Proc. of the OECD–NEA Specialists' Meeting on Accelerator–Based Transmutation, PSI Würenlingen/Villigen, Switzerland, 24–26 March 1992

[9] T. Nishida et al., "Benchmark Study on the Computational Model in the Accelerator–Based Transmutation Simulation Code", Proc. of the OECD–NEA Specialists' Meeting on Accelerator–Based Transmutation, PSI Würenlingen/Villigen, Switzerland, 24–26 March 1992

[10] T.W. Armstrong et al., "A Comparison of High–Energy Fission Models", Nucl. Inst. and Methods in Physics Research, **222**, 540–547 (1984)

Table 1. **Fission-to-Capture ratios for TRU nuclides with $T_{1/2} > 10$ a**

	D_2O CELL	LWR (KKG)	FBR (SPX)	MOLTEN SALT	PHOENIX REF.	PHOENIX METAL
^{237}Np	0.00	0.02	0.23	0.45	0.94	1.10
^{238}Pu	0.03	0.09	2.16	3.39	5.94	6.94
^{239}Pu	2.27	1.76	3.69	5.27	8.98	10.26
^{240}Pu	0.00	0.00	0.74	1.38	2.88	3.35
^{242}Pu	0.00	0.02	0.61	1.26	2.68	3.19
^{244}Pu	0.02	0.16	1.10	2.60	8.44	10.42
^{241}Am	0.01	0.01	0.15	0.30	0.57	0.63
^{242m}Am	4.88	4.93	6.55	7.03	7.83	8.22
^{243}Am	0.00	0.01	0.14	0.29	0.59	0.66
^{243}Cm	4.99	5.88	7.42	9.97	24.44	33.46
^{244}Cm	0.04	0.06	0.84	1.51	2.82	3.22
^{245}Cm	6.58	6.87	6.24	7.79	12.96	14.96
^{246}Cm	0.07	0.22	1.27	2.69	5.77	6.41
^{247}Cm	1.47	1.56	6.40	9.01	13.60	15.56
^{248}Cm	0.05	0.12	1.40	2.74	6.10	6.98

Table 2. " **Fuel neutron production " for TRU nuclides with $T_{1/2} > 10$ a**

Flux	D_2O CELL 1E16	LWR (KKG) 1E14	FBR (SPX) 1E15	MOLTEN SALT 1E15	PHOENIX REF. 1E15	PHOENIX METAL 1E15
^{237}Np	0.20	-1.05	0.67	1.03	1.32	1.43
^{238}Pu	0.07	-0.10	1.41	1.65	1.83	1.88
^{239}Pu	1.01	0.72	1.53	1.74	1.89	1.93
^{240}Pu	0.04	-0.30	1.00	1.41	1.73	1.82
^{242}Pu	-0.56	-1.16	0.60	1.27	1.70	1.83
^{244}Pu	1.38	1.55	1.94	2.14	2.25	2.26
^{241}Am	-0.43	-0.94	0.68	1.13	1.52	1.68
^{242m}Am	1.73	1.63	1.89	2.00	2.10	2.15
^{243}Am	0.39	-0.22	0.71	1.19	1.59	1.75
^{243}Cm	2.06	1.90	2.12	2.23	2.34	2.38
^{244}Cm	1.39	0.76	1.47	1.80	2.06	2.13
^{245}Cm	2.36	2.43	2.63	2.76	2.90	2.95
^{246}Cm	0.33	0.75	2.23	2.58	2.76	2.79
^{247}Cm	1.18	1.31	2.41	2.59	2.70	2.74
^{248}Cm	0.11	0.31	1.68	2.18	2.55	2.64

Table 3. **Summary of the interactions in a Phoenix module (per incident particle)**

Parameters	HEF (enabled) $k_{eff} = 0.796$	HEF (disabled) $k_{eff} = 0.796$
Number of High-Energy[2] Leakage Neutrons	1.5 (102MeV)[4]	1.5 (103MeV)[4]
Number of Low-Energy[3] Leakage Neutrons	47.1	46.1
Total Number of Leakage Neutrons	48.6 (34%)[5]	47.6 (34%)[5]
Number of Low-Energy Source Neutrons	25.3 (4.08MeV)[4]	26.0 (3.73MeV)[4]
Total Number of Neutrons Produced by HET	26.8	27.5
Number of High-Energy Fissions	2.06	0.00
Number of Low-Energy Fissions	35.29	34.32
Total Number of Fissions	37.35	34.32
Total Number of Evaporations	5.2	7.3
Number of Low-Energy Captures	51.7	51.0
Total Number of Transmutations	94.2	92.7
Power Deposited by High-Energy Interactions	1.40	0.92
Power Deposited by Low-Energy Interactions	7.59	7.38
Target Power	8.99	8.30
(in MW_{th}/mA)		

---

[2] > 15 MeV

[3] < 15 MeV

[4] mean energy of the particle

[5] proportion of the total neutrons produced leaking out

Table 4. **Sensitivity of fission-to-capture ratios for TRU nuclides with $T_{1/2} > 10$ a**

	CRIT.	k=0.8	NOFIS.	REF.	ENDFB-6	JENDL-3
^{237}Np	0.61	0.88	0.92	0.94	1.00	0.98
^{238}Pu	4.46	5.71	5.88	5.94	6.14	5.97
^{239}Pu	7.32	8.74	8.91	8.98	9.16	9.02
^{240}Pu	1.95	2.71	2.84	2.88	3.02	2.91
^{242}Pu	1.76	2.51	2.64	2.68	2.82	2.72
^{244}Pu	5.26	7.87	8.29	8.44	8.89	8.58
^{241}Am	0.37	0.53	0.56	0.57	0.61	0.58
^{242m}Am	7.00	7.67	7.79	7.83	7.98	7.85
^{243}Am	0.37	0.54	0.58	0.59	0.62	0.60
^{243}Cm	19.19	23.99	24.28	24.44	24.74	24.75
^{244}Cm	1.90	2.65	2.78	2.82	2.97	2.86
^{245}Cm	10.87	12.68	12.88	12.96	13.18	13.06
^{246}Cm	3.76	5.35	5.67	5.77	6.10	5.88
^{247}Cm	11.08	13.26	13.51	13.60	13.89	13.64
^{248}Cm	3.99	5.66	6.00	6.10	6.41	6.19

Table 5. **Sensitivity of 'fuel neutron production' for TRU nuclides with $T_{1/2} > 10$ a**

Flux	CRIT. 1E15	k=0.8 1E15	NOFIS. 1E15	REF. 1E15	ENDFB-6 1E15	JENDL-3 1E15
^{237}Np	1.12	1.27	1.31	1.32	1.39	1.32
^{238}Pu	1.71	1.79	1.82	1.83	1.84	1.83
^{239}Pu	1.81	1.86	1.89	1.89	1.90	1.89
^{240}Pu	1.52	1.67	1.72	1.73	1.75	1.73
^{242}Pu	1.42	1.63	1.69	1.70	1.73	1.71
^{244}Pu	2.16	2.20	2.24	2.25	2.26	2.25
^{241}Am	1.24	1.45	1.51	1.52	1.56	1.54
^{242m}Am	2.01	2.08	2.10	2.10	2.11	2.11
^{243}Am	1.29	1.51	1.58	1.59	1.63	1.61
^{243}Cm	2.28	2.32	2.34	2.34	2.35	2.35
^{244}Cm	1.87	2.00	2.05	2.06	2.08	2.06
^{245}Cm	2.82	2.87	2.89	2.90	2.91	2.90
^{246}Cm	2.60	2.69	2.75	2.76	2.77	2.77
^{247}Cm	2.61	2.67	2.70	2.70	2.71	2.71
^{248}Cm	2.31	2.47	2.54	2.55	2.57	2.56

Table 6. **Mass changes in kg/year for the Phoenix concept from the below 15 MeV processes**

Nuclides	Initial Mass Inventory (kg) (per module)	PHOENIX/BNL REF. 450 MWth (per module)	PHOENIX/BNL (w/o fission) 450 MWth (per module)
^{234}U		0.51	0.51
^{237}Np	1369	- 117	- 116
^{238}Np		0.32	0.32
^{238}Pu		96.0	95.6
^{239}Pu		1.09	1.15
^{240}Pu		2.47	2.35
^{241}Pu		0.03	0.03
^{242}Pu		15.1	15.1
^{241}Am	1593	- 165	- 165
^{242}Am		0.14	0.14
^{242m}Am		14.5	14.4
^{243}Am	289	- 23.7	- 23.5
^{242}Cm		25.9	25.8
^{243}Cm		0.35	0.36
^{244}Cm	57	+ 7.80	+ 7.91
^{245}Cm		1.15	1.15
Total	3308	- 140	- 140
k_{boc}		0.796	0.796
Δk		0.019	0.021
LWR Support Ratio		8.7	8.6

Table 7. **Mass changes in kg/year for the Phoenix concept due to high-energy processes only**

Nuclides	Initial Mass Inventory (kg) (per module)	PHOENIX/BNL REF. 450 MWth (per module)	PHOENIX/BNL (w/o fission) 450 MWth (per module)
^{234}U		0.02	0.05
^{237}Np	1369	- 4.05	- 4.43
^{238}Np		0.00	0.00
^{238}Pu		0.01	0.05
^{239}Pu		0.03	0.07
^{240}Pu		0.10	0.13
^{241}Pu		0.03	0.03
^{242}Pu		0.01	0.02
^{241}Am	1593	- 4.73	- 5.07
^{242}Am		0.05	0.08
^{242m}Am		–	–
^{243}Am	289	- 0.85	- 0.94
^{242}Cm		0.01	0.01
^{243}Cm		0.01	0.02
^{244}Cm	57	- 0.19	- 0.19
^{245}Cm		–	–
Total	3308		

Figure 1. Sensitivity of the neutron source spectrum

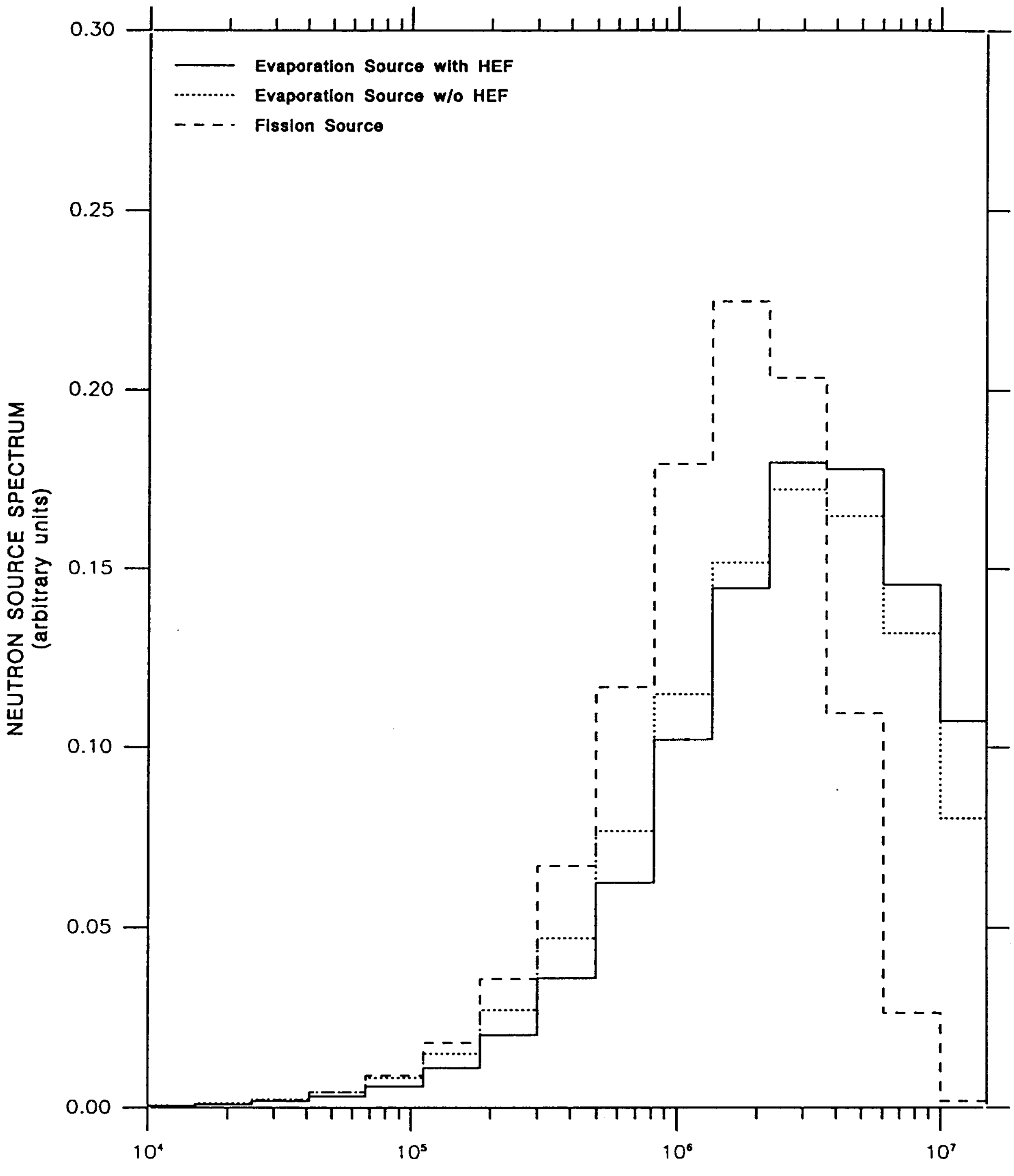

Figure 2. Mass distribution of the high-energy reaction products

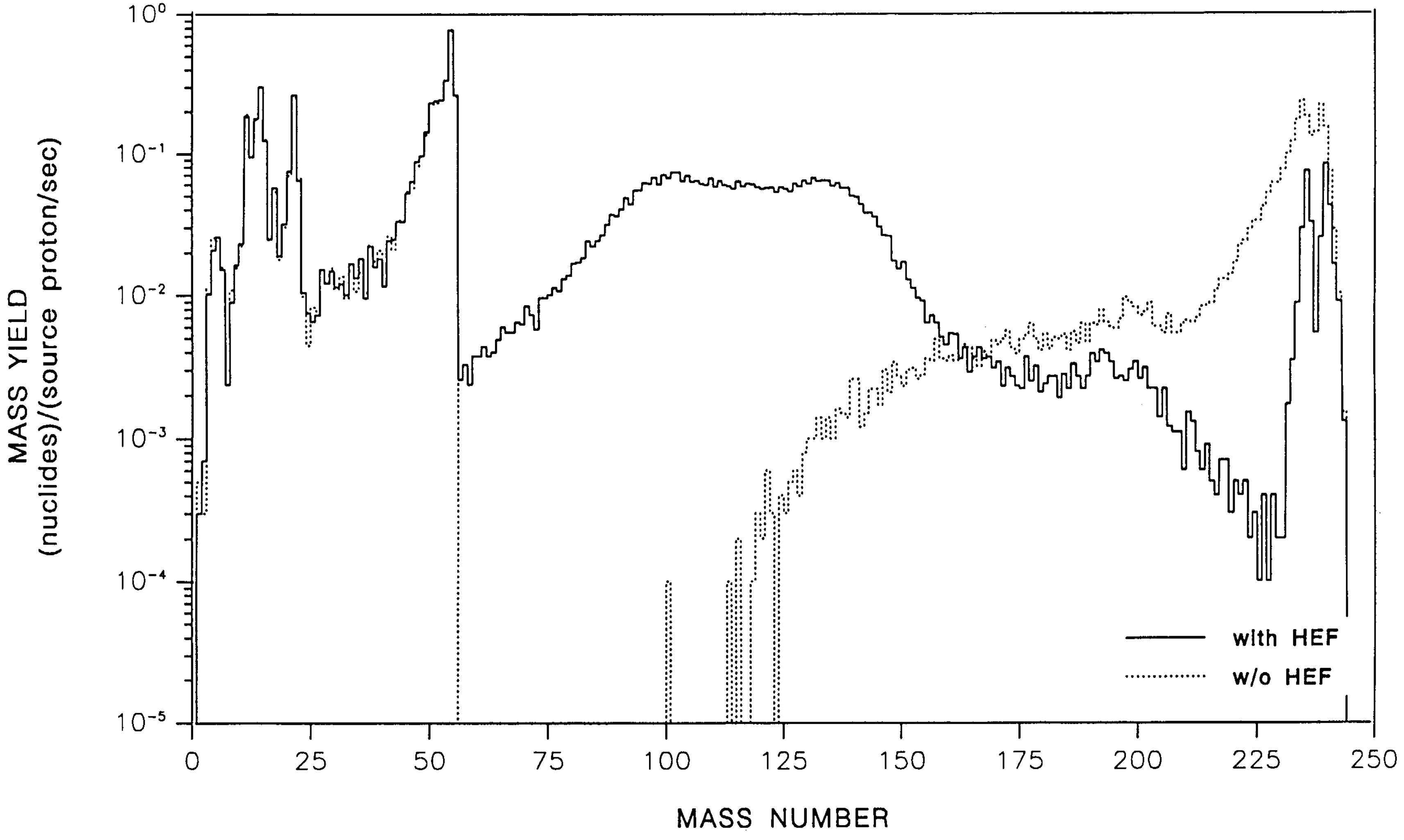

Figure 3. Toxicity of the high-energy reaction products (per mA)

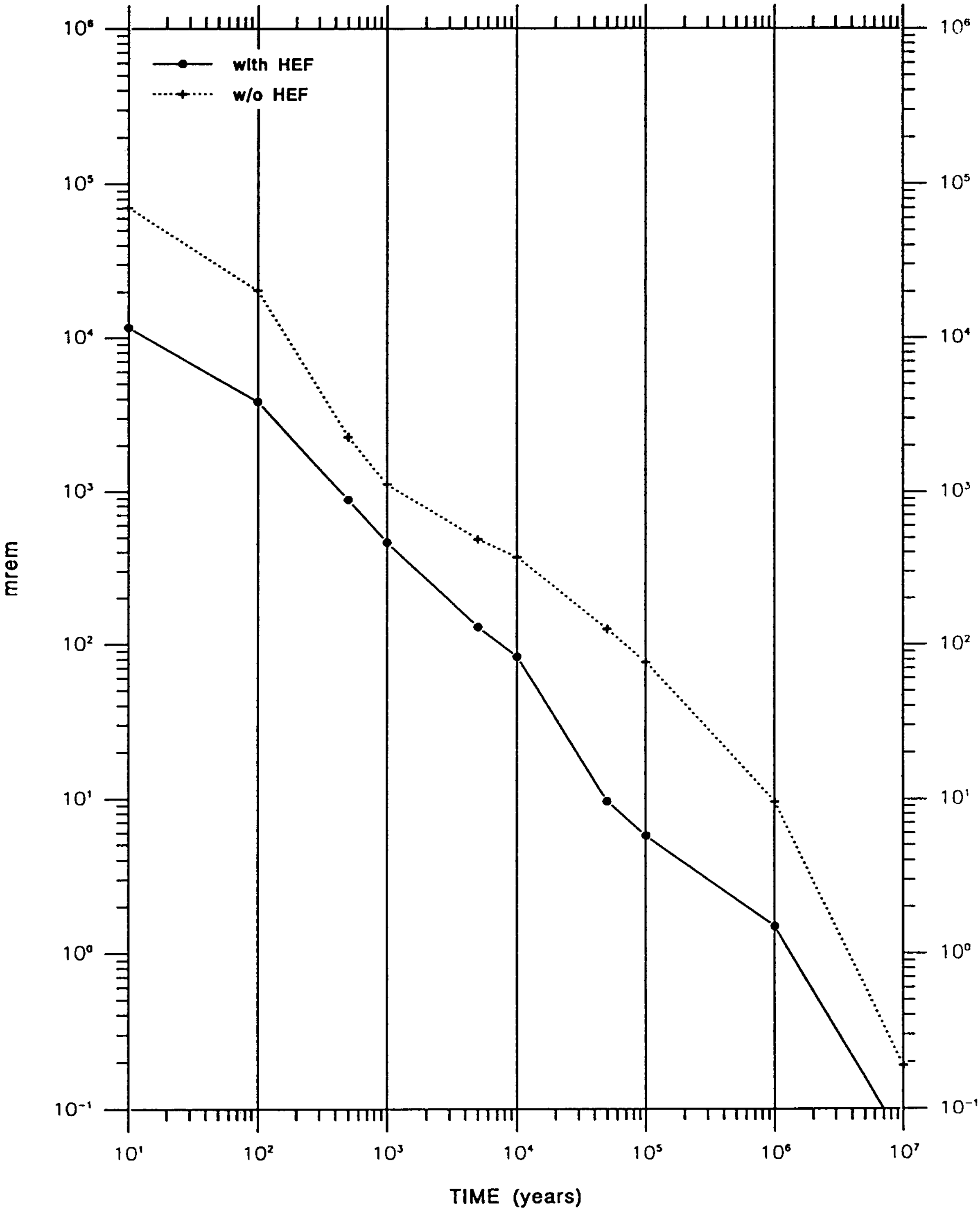

Figure 4. Sensitivity of the neutron flux spectrum