A PHYSICAL ASSESSMENT OF THE EFFECTIVENESS OF DIFFERENT ACTINIDE TRANSMUTATION SYSTEMS

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

ABSTRACT

A general method for investigating the effectiveness of actinide transmutation systems is proposed. The method allows to assess the impact of different long–term transmutation strategies on the actinide inventories of the systems, the composition of the actinide waste, and the radiological risk associated with the disposal of this waste. In a comparative study, the method is applied to systems with a wide range of characteristics including a PWR, a fast reactor, a high–flux superthermal system, and two accelerator–based systems with fast neutron spectra.

The results of the study emphasise the importance of a good overall neutron balance for completely burning the actinides. As to the radiological risk of the waste, it is found that most transuranic actinides can be recycled in fast reactors and PWRs with similar risks, and that high–flux superthermal systems burn actinides generally with a somewhat smaller risk than other systems. Interestingly, it appears that, in the long range, the radiological risk of the waste cannot be reduced by changing from the uranium–plutonium to the thorium–uranium fuel cycle.

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

INTRODUCTION

Actinides contribute significantly to the long–term toxicity of radioactive wastes, since they are all unstable and decay through extended chains, involving long–lived α emmitters, to nuclides in the stable range. The decays can be by–passed by fissioning the actinides. For a long–term nuclear energy system, the goal must be therefore to fission the actinides with a high overall efficiency. A transmutation system can be termed to be "effective", if it contributes to achieve this goal. This means, for instance, that the fissionable nuclides, i.e. the even–neutron nuclides with a high–energy fission threshold, should not be treated as waste, but utilised more extensively for energy production.

The effectiveness of transmutation systems is usually assessed by means of detailed calculations of the mass flows for the specific concepts and fuel management schemes. This approach was adopted, for example, in the "Overview of Physics Aspects of Different Transmutation Concepts" prepared by a task force of the OECD–NEA Nuclear Science Committee [1]. While yielding all parameters of interest, the detailed analysis of a system usually requires a significant computational effort. This effort can become prohibitive, if many systems are to be compared using consistent assumptions and approximations.

For this reason and to improve the understanding of the basic phenomena, there is a need for simplified approaches which concentrate on specific aspects of the systems. Transmutation properties have been discussed, for instance, in terms of the fission–to–capture ratios of the nuclides to be burnt or the overall neutron balance of the systems. For the investigation of long–term transmutation strategies, the present paper proposes a method which allows to quantify the impact of the strategies on the actinide inventories of the systems, the composition of the actinide waste, and the radiological risk associated with the disposal of this waste. The method is used to compare the transmutation effectiveness of systems with different characteristics and different fuel cycles on a consistent basis, i.e. using a common data base.

COMPARED SYSTEMS

The comparison includes the following systems:

- A normal PWR
  The fuel cell characteristics are those of the 920 MWe plant of Gösgen, and the average neutron flux is assumed to be 10^{14} $cm^{-2}s^{-1}$.

- A MOX–fuelled fast reactor (FR)
  The composition of the fuel corresponds to that of the fresh core of Superphénix, and the average flux is assumed to be 10^{15} $cm^{-2}s^{-1}$.

- A "superthermal" system with a high flux (STS)
  A well–moderated thermal neutron spectrum, typical for the D_2O moderator of a continuous spallation neutron source, is assumed. The system simulates the base–case design of the Los Alamos ATW concept [2] with a neutron flux of 10^{16} $cm^{-2}s^{-1}$.

- An accelerator–driven "fast" molten salt system (FMS)
  This is a directly driven system with chloride molten salt fuel and a continuous reprocessing system for removing the fission products. The geometry and the composition are those of the "molten salt core system" proposed by JAERI [3], and the average neutron flux is 10^{15} $cm^{-2}s^{-1}$. The neutrons have a "harder" spectrum (i.e. a higher mean energy) than the neutrons in a MOX–fuelled fast reactor.
- An accelerator–driven "superfast" system using metal fuel (SFS)
  Here, the idea is to combine the favourable neutronics of an actinide target and a metal fuel cycle with low reprocessing losses. The system features a sodium–cooled target with actinide–zirconium fuel and full actinide recycling. The geometry conforms with that of the Phoenix concept [4], and the neutron flux is the same as that assumed for the other fast systems. Of the investigated systems, this system has the hardest neutron spectrum.

MODELLING OF THE SYSTEMS

The parameters used in this investigation are derived from a common set of spatially averaged one–group cross sections prepared as follows:

The cross sections for the PWR and the superthermal system were obtained from thermal cell calculations using the BOXER code [5] together with a cross section library derived from the JEF–1 evaluated nuclear data file.

The cross sections for the fast reactor and the hybrid systems were generated using two–dimensional cylindrical models of the systems. With the height of the cylinder fixed to represent the height of the reactor or the depth of the target, the radius was adjusted to give the appropriate beginning–of–life multiplication factor. For the subcritical cases, neutron sources were calculated using a version of the HETC code which includes high–energy fission [6]. Neutron spectrum calculations below 15 MeV are based on JEF–2.2 and the computational scheme described in Ref. 7.

The sensitivity of the parameters to the data and the modelling of the systems is discussed in Ref. 8.

FISSION–TO–CAPTURE RATIOS

In all fission–based systems, neutron–induced fission compete with capture reactions. In general, neutron captures are undesirable, since they lead to an increase rather than a reduction of the atomic mass number. It can therefore be speculated that the transmutation effectiveness of a system for a particular actinide increases with the fission–to–capture ratio of this actinide.

Table 1 shows that, for most actinides, fast neutron spectra yield a higher fission–to–capture ratio than thermal spectra. In accelerator–based fast systems, the mean neutron energy and thus the fission–to–capture ratios increase beyond the limits of normal fast reactors due to the subcriticality of the target and the effect of the evaporation neutrons which have higher energies than the fission neutrons. As expected, the fissionable nuclides show particularly strong spectrum hardening effects. There may therefore be an incentive for designing accelerator–based fast systems specifically with the aim of fissioning the fissionable nuclides.

OVERALL NEUTRON BALANCE

An 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 while maintaining criticality and taking account of losses due to neutrons leaking out of the system. In Ref. 9, Salvatores et al. have proposed to measure the overall neutron balance in terms of the "neutron production" parameter, –D. Unlike other neutron balance parameters, the "neutron production" depends on the ratio of neutron induced reactions (fissions, captures, n,2n reactions) to radioactive decays and therefore on the neutron flux.

Table 1: Fission–to–Capture Ratios for Actinides with $T_{1/2} > 10$ a

Nuclide	STS	PWR	FR	FMS	SFS
^{230}Th	0.00	0.00	0.20	0.43	1.87
^{232}Th	0.00	0.01	0.03	0.06	0.16
^{231}Pa	0.00	0.01	0.09	0.18	0.52
^{232}U	1.13	1.49	3.45	4.79	9.58
^{233}U	10.21	7.91	10.68	11.84	15.29
^{234}U	0.00	0.03	0.61	1.12	2.60
^{235}U	5.57	4.23	3.61	4.29	6.39
^{236}U	0.01	0.04	0.21	0.42	0.95
^{238}U	0.00	0.13	0.18	0.33	0.99
^{237}Np	0.00	0.02	0.23	0.45	1.32
^{238}Pu	0.03	0.09	2.16	3.39	7.93
^{239}Pu	2.27	1.76	3.69	5.27	11.35
^{240}Pu	0.00	0.00	0.74	1.38	3.96
^{241}Pu	2.91	2.95	4.67	4.90	5.34
^{242}Pu	0.00	0.02	0.61	1.26	3.82
^{244}Pu	0.02	0.16	1.10	2.60	12.87
^{241}Am	0.01	0.01	0.15	0.30	0.75
^{242m}Am	4.88	4.93	6.55	7.03	8.74
^{243}Am	0.00	0.01	0.14	0.29	0.81
^{243}Cm	4.99	5.88	7.42	9.97	39.48
^{244}Cm	0.04	0.06	0.84	1.51	3.83
^{245}Cm	6.58	6.87	6.24	7.79	16.54
^{246}Cm	0.07	0.22	1.27	2.69	7.62
^{247}Cm	1.47	1.56	6.40	9.01	17.26
^{248}Cm	0.05	0.12	1.40	2.74	8.30

Nuclide–dependent "neutron production" values for the different systems are shown in Table 2. For the most abundant minor actinides in LWR–discharged fuels (^{237}Np, ^{241}Am, ^{243}Am) the overall neutron balance in a PWR is negative, indicating that the chain of successive transmutations does not provide enough neutrons to support itself. In a superthermal system, the overall neutron balance improves but remains poor compared with that of a fast reactor. Superfast systems offer the possibility of enhancing the overall neutron balance beyond the limit of fast reactors. This can be desirable when the system is intended to transmute not only actinides, but also fission products.

ASYMPTOTIC ACTINIDE INVENTORIES

Nuclide inventories and fuel compositions play an important role in system studies and risk analyses: the former have an impact on the overall characteristics of the systems and the core accident risk; the latter influence the different types of risks arising from the leakage and migration of radiation and activity in the fuel cycle, including the long–term risk associated with waste repositories. From a safety viewpoint, small nuclide inventories per MW are advantageous, since they comply with the goal of a small overall risk.

Table 2: Overall Neutron Balance for Actinides with $T_{1/2} > 10$ a (–D in Ref. 9)

Nuclide	STS	PWR	FR	FMS	SFS
^{230}Th	-0.75	-0.78	-0.07	0.25	0.77
^{232}Th	-1.21	0.17	0.38	0.55	0.74
^{231}Pa	0.25	0.22	0.67	0.83	1.08
^{232}U	1.25	1.20	1.60	1.70	1.85
^{233}U	1.26	1.18	1.35	1.42	1.50
^{234}U	-0.17	-0.34	0.55	0.92	1.26
^{235}U	0.82	0.61	0.94	1.13	1.35
^{236}U	-1.62	-1.92	0.02	0.54	1.06
^{238}U	-0.14	-0.04	0.73	1.06	1.43
^{237}Np	0.20	-1.05	0.67	1.03	1.43
^{238}Pu	0.07	-0.10	1.41	1.65	1.88
^{239}Pu	1.01	0.72	1.53	1.74	1.93
^{240}Pu	0.04	-0.30	1.00	1.41	1.82
^{241}Pu	1.04	0.70	1.26	1.50	1.79
^{242}Pu	-0.56	-1.16	0.60	1.27	1.83
^{244}Pu	1.38	1.55	1.94	2.14	2.26
^{241}Am	-0.43	-0.94	0.68	1.13	1.68
^{242m}Am	1.73	1.63	1.89	2.00	2.15
^{243}Am	0.39	-0.22	0.71	1.19	1.75
^{243}Cm	2.06	1.90	2.12	2.23	2.38
^{244}Cm	1.39	0.76	1.47	1.80	2.13
^{245}Cm	2.36	2.43	2.63	2.76	2.95
^{246}Cm	0.33	0.75	2.23	2.58	2.79
^{247}Cm	1.18	1.31	2.41	2.59	2.74
^{248}Cm	0.11	0.31	1.68	2.18	2.64

Considering that the ultimate goal is to incorporate transmutation into closed long–term nuclear energy systems, it is useful to calculate "asymptotic actinide inventories", i.e. actinide inventories resulting from the continuous burning of individual nuclides in the systems of interest assuming that the nuclides are exposed to a fixed neutron spectrum and flux and burnt until all nuclear reactions are in equilibrium. The asymptotic inventory for burning a mixture of nuclides can be obtained easily by linearly combining the nuclide–specific inventories.

A direct method has been used to efficiently calculate such inventories. In brief, the asymptotic inventory of nuclide i resulting from burning nuclide j, $N_{i,j}$, is obtained from the equation

$$\sum_k P_{i,j,k} = \lambda_{i,eff} N_{i,j} \ ,$$

where P and λ are the production rate and the effective half–life for the given neutron spectrum and flux. The production rate is summed over the generations, k, of the chain of transmutations until the initial nuclei are fissioned (a small fraction of the initial nuclei is "lost", i.e. transmuted to heavy nuclei with atomic numbers outside the range of the calculation, $90 < Z < 96$, but for the important actinides this fraction is negligible). The method is much faster than a normal burnup calculation, allowing asymtotic inventories to be calculated accurately for a wide range of parameters.

For nuclides with $T_{1/2} > 10$ a, asymptotic inventories resulting from burning ^{237}Np and ^{239}Pu are given in Tables 3 and 4. The k_{inf}[1] values in the tables indicate to what extent the asymptotic actinide mixtures can themselves maintain criticality. Obviously, burning ^{237}Np in a PWR does not produce a critical inventory. In this case, fissile material has to be added to the mixture whereby the effective inventory is increased, or the system has to be driven using externally generated neutrons.

Comparing the PWR with the fast reactor, it can be seen that, for the more abundant actinides, the asymptotic inventories of the latter are higher. However, the fast reactor has lower inventories of the higher plutonium isotopes, ^{243}Am, and the curium isotopes.

Interestingly, it appears that increasing the mean neutron energy beyond that of a fast reactor further enhances the inventory of the nuclide being burnt. On the other hand, hardening the neutron spectrum has the expected effect of reducing americium and curium inventories as well as ^{238}Pu and ^{240}Pu build–up from ^{237}Np and ^{239}Pu, respectively.

Table 3: Asymptotic Inventories Resulting from Burning ^{237}Np
(No. of atoms normalised to 1 fission/s)

Nuclide	STS	PWR	FR	FMS	SFS
^{230}Th	1.17E-05	1.80E+03	5.24E+04	5.06E+04	3.95E+04
^{232}Th	7.22E-07	6.34E+01	5.42E+01	4.87E+01	3.11E+01
^{231}Pa	2.64E-06	8.42E+02	2.81E+03	3.49E+03	3.19E+03
^{232}U	2.97E-03	4.01E+03	2.90E+04	2.38E+05	3.22E+05
^{233}U	1.73E-03	1.87E+03	2.06E+04	1.32E+05	1.03E+05
^{234}U	4.21E+01	4.21E+07	1.17E+08	1.03E+08	5.84E+07
^{235}U	8.51E+00	2.04E+07	2.82E+07	2.24E+07	1.05E+07
^{236}U	4.34E+01	1.89E+07	2.30E+07	1.64E+07	7.13E+06
^{238}U	3.79E+01	4.43E+05	1.63E+04	5.81E+03	9.30E+02
^{237}Np	9.29E+05	3.30E+08	5.47E+08	6.59E+08	8.04E+08
^{238}Pu	1.08E+05	3.52E+08	4.35E+08	3.72E+08	2.27E+08
^{239}Pu	5.13E+04	6.03E+07	9.95E+07	6.63E+07	2.30E+07
^{240}Pu	2.74E+04	1.65E+07	5.29E+07	2.39E+07	3.55E+06
^{241}Pu	1.40E+04	2.45E+07	6.29E+06	2.23E+06	1.87E+05
^{242}Pu	6.95E+04	2.99E+07	6.16E+06	1.87E+06	1.12E+05
^{244}Pu	8.40E+03	8.10E+03	6.03E+01	7.86E+00	9.27E-02
^{241}Am	5.68E+00	3.70E+06	4.35E+06	1.83E+06	1.88E+05
^{242m}Am	7.63E-02	9.56E+04	3.50E+05	1.28E+05	9.98E+03
^{243}Am	3.02E+04	1.78E+07	1.61E+06	4.24E+05	1.74E+04
^{243}Cm	6.30E-01	2.18E+04	1.17E+04	3.19E+03	8.93E+01
^{244}Cm	1.04E+05	2.76E+07	1.21E+06	2.23E+05	4.68E+03
^{245}Cm	2.18E+03	3.78E+06	1.24E+05	2.58E+04	5.29E+02
^{246}Cm	9.14E+04	1.67E+07	1.71E+05	1.72E+04	8.30E+01
^{247}Cm	2.88E+03	1.18E+06	1.75E+04	1.25E+03	3.33E+00
^{248}Cm	1.54E+04	2.64E+06	9.42E+03	4.22E+02	4.91E-01
k_{inf}	1.077	0.735	1.299	1.535	1.958

[1] Neutron multiplication factor of an infinite system

When applying these results to practical systems, it has to be taken into account that the asymptotic inventories depend on the neutron flux. Typical fluxes in PWRs and fast systems are 2 to 3 times higher than those assumed in the present comparison. On the other hand, the fuel cycle is not explicitly modelled in the calculations. Assuming a batch–type processing, the fluxes were therefore appropriately scaled to simulate the out–of–pile time of the fuel.

Compared with the PWR, the superthermal system has lower inventories than one would expect from the flux ratio alone. This is due to the well–moderated neutron spectrum and the fact that short–lived capture products with a high fission cross section are fissioned before they decay. The spectrum effect is relatively important and reduces the inventories of the nuclides being burnt by additional factors of 3.6 and 4.5 for ^{237}Np and ^{239}Pu, respectively. In the case of ^{237}Np, the ^{238}Np fission effect mitigates considerably the build–up of ^{238}Pu (cf. ^{238}Pu / ^{237}Np ratios in Table 3).

Table 4: Asymptotic Inventories Resulting from Burning ^{239}Pu
(No. of atoms normalised to 1 fission/s)

Nuclide	STS	PWR	FR	FMS	SFS
^{230}Th	1.77E-10	5.45E+01	1.65E+03	1.10E+03	4.21E+02
^{232}Th	4.25E-08	2.22E+00	4.80E+00	4.30E+00	3.08E+00
^{231}Pa	4.12E-11	2.55E+01	8.82E+01	5.57E+01	1.19E+01
^{232}U	3.71E-10	4.39E+01	1.35E+02	3.44E+02	1.41E+02
^{233}U	7.23E-10	2.18E+01	4.28E+02	1.98E+03	7.00E+02
^{234}U	6.38E-04	1.28E+06	3.70E+06	2.24E+06	6.26E+05
^{235}U	4.65E-02	6.35E+05	1.06E+06	7.17E+05	4.49E+05
^{236}U	2.56E+00	6.62E+05	2.04E+06	1.45E+06	7.06E+05
^{238}U	2.37E+00	3.69E+04	6.24E+03	2.98E+03	7.17E+02
^{237}Np	9.41E-02	2.53E+05	1.12E+06	8.41E+05	3.38E+05
^{238}Pu	1.64E+00	1.07E+07	1.37E+07	8.11E+06	2.42E+06
^{239}Pu	1.62E+05	7.25E+07	4.39E+08	4.88E+08	5.51E+08
^{240}Pu	8.18E+04	1.98E+07	2.33E+08	1.76E+08	8.50E+07
^{241}Pu	4.18E+04	2.95E+07	2.77E+07	1.64E+07	4.47E+06
^{242}Pu	2.08E+05	3.60E+07	2.72E+07	1.37E+07	2.69E+06
^{244}Pu	2.51E+04	9.75E+03	2.66E+02	5.78E+01	2.22E+00
^{241}Am	1.70E+01	4.45E+06	1.92E+07	1.34E+07	4.51E+06
^{242m}Am	2.28E-01	1.15E+05	1.54E+06	9.44E+05	2.39E+05
^{243}Am	9.02E+04	2.15E+07	7.09E+06	3.12E+06	4.16E+05
^{243}Cm	1.88E+00	2.62E+04	5.14E+04	2.35E+04	2.14E+03
^{244}Cm	3.11E+05	3.33E+07	5.32E+06	1.64E+06	1.12E+05
^{245}Cm	6.53E+03	4.54E+06	5.46E+05	1.90E+05	1.27E+04
^{246}Cm	2.73E+05	2.01E+07	7.55E+05	1.27E+05	1.99E+03
^{247}Cm	8.60E+03	1.42E+06	7.71E+04	9.20E+03	7.98E+01
^{248}Cm	4.61E+04	3.17E+06	4.16E+04	3.10E+03	1.18E+01
k_{inf}	1.518	1.326	2.069	2.372	2.756

KEY NUCLIDES FOR RADIOLOGICAL RISK ASSESSMENT

Actinides which contribute to the radiological risk of radioactive waste repositories are actinides with lifetimes greater than about 10^4 a and their α active daughter products. Examples are ^{237}Np and its daughter product ^{229}Th, ^{226}Ra, a daughter product of ^{234}U, and ^{231}Pa, a daughter product of ^{235}U. Risk analyses have shown that, due to its low solubility and strong sorption, plutonium does not directly contribute to the risk. Risk contributions from extremely long–lived and therefore practically inactive nuclides as ^{238}U ($T_{1/2} = 4.5 \cdot 10^9$ a) and ^{232}Th ($T_{1/2} = 1.4 \cdot 10^{10}$ a) are small or negligible.

It is evident that the risk of waste repositories is closely related to the activity of one or two long–lived "key nuclides" – mainly uranium isotopes – from each of the four principal actinide decay chains. In general, the precursors of these nuclides have already decayed when the nuclides or their daughter products enter the biosphere. Assuming that all daugther nuclides are shorter–lived, the resulting dose or risk can be expected to be proportional to the activity of the nuclide and to an effective dose conversion factor including daughter contributions for equilibrium conditions.

A practical implementation of this concept is shown in Table 5. For each key nuclide, the table specifies the precursors contributing to its activity, an effective dose conversion factor, and nuclides which produce dominant dose contributions. The effective dose conversion factor is derived from ICRP–61 [10] assuming that all daughter products are in equilibrium and contribute equally to the dose (more realistic models for calculating effective dose conversion factors are being investigated).

Table 5: Key Nuclides for Radiological Risk Assessment

Principal decay chain	Key nuclide	$T_{1/2}$ [a]	Included precursors	Eff. dose factor [Sv/Bq]	Principal dose contributors
4n	^{236}U	$2.3 \cdot 10^7$	all	$3.0 \cdot 10^{-8}$	^{236}U
4n + 1	^{237}Np	$2.1 \cdot 10^6$	all	$1.2 \cdot 10^{-6}$	^{237}Np, ^{229}Th
4n + 1	^{233}U	$1.6 \cdot 10^5$	^{233}Pa	$5.4 \cdot 10^{-7}$	^{229}Th
4n + 2	^{234}U	$2.4 \cdot 10^5$	all from ^{238}Pu branch	$3.2 \cdot 10^{-7}$	^{226}Ra
4n + 2	^{238}U	$4.5 \cdot 10^9$	all	$3.4 \cdot 10^{-7}$	^{226}Ra
4n + 3	^{235}U	$7.8 \cdot 10^8$	all	$4.2 \cdot 10^{-6}$	^{231}Pa, ^{227}Ac[1]
4n + 3	^{231}Pa	$3.3 \cdot 10^4$	none	$4.2 \cdot 10^{-6}$	^{231}Pa, ^{227}Ac[1]

[1] Daughter products of ^{227}Ac

For the thorium chain (4n chain), ^{236}U is chosen as a key nuclide[1]. For systems based on the uranium–plutonium fuel cycle, the key nuclide of the neptunium chain (4n+1 chain) is ^{237}Np. ^{233}U is included, because it is important in thorium–based systems. With regard to its abundance in uranium–based fuels, ^{238}U is included as a second nuclide of the radium chain (4n+2 chain). Lastly, ^{231}Pa is included as a second nuclide of the actinium chain (4n+3 chain), since it is produced in significant amounts in thorium–based systems and its specific activity is much higher than that of ^{235}U.

APPLICATION OF SCHEME TO SYSTEMS WITH BATCH–TYPE PROCESSING

The actinide inventory of the waste repository per fission may be written as

$$F_i \tilde{N}_{i,j} \ ,$$

where F_i and $\tilde{N}_{i,j}$ are the number of heavy atoms in the waste stream relative to the number of fissions and the fuel composition, respectively. The latter can be obtained by normalising the asymptotic actinide inventory, viz.,

$$\tilde{N}_{i,j} = \frac{N_{i,j}}{\sum_i N_{i,j}} \ .$$

The quantity F_i – in the following to be termed "waste fraction" – depends on the burnup of the fuel and the reprocessing and fuel fabrication losses. For a batch–type processing, it is obtained from the expression

$$\frac{L_i(1-B)}{B} \ ,$$

where the burnup, B, and the nuclide–dependent losses, L_i, have to be given as fractions of the inventory.

For a waste fraction of 1.0, the scheme of the key nuclides yields the activities in Tables 6 and 7. The tables contain all activities greater than one hundredth of the dominating activity. By multiplying the activities with the appropriate effective dose conversion factors from Table 5, one obtains nuclide–specific risks. Depending on the characteristics of the waste repository, these risks are independent or cumulative. Nuclide–specific and cumulative (total) risks for burning ^{232}Th, ^{238}U, ^{237}Np and ^{239}Pu are compiled in Table 8. Burnup and losses can be accounted for by multiplying the values in the tables with the appropriate waste fractions.

ACTINIDE RECYCLING IN THERMAL AND FAST SYSTEMS

For LWRs and fast reactors, the achievable burnup, expressed in atom % of the fissile material, is about the same. If the systems use the same type of fuel cycle, it can be assumed that they also have similar reprocessing and fuel fabrication losses. In this case, the waste fractions are comparable and the numbers in Tables 6 to 8 can be used directly to assess relative activities and risks.

The suitability of LWRs and fast systems for utilising and burning plutonium, neptunium and americium may be discussed using the cumulative risk ratios in Table 9. The FR/PWR ratios show that the cumulative nuclide–specific risks for the fast reactor and the PWR lie within a factor of two, with two exceptions: ^{239}Pu burns cleaner in the fast, and ^{242m}Am burns significantly cleaner in the thermal neutron spectrum. It is noteworthy that this applies to both fissile and fissionable nuclides. As outlined before, thermal systems do, however, not provide an adequate overall neutron balance for completely burning fissionable nuclides, necessitating the use of additonal fuel which augments the risk, or a hybrid system.

[1] Due to the inclusion of the longer–lived ^{244}Pu ($T_{1/2} = 8.3 \cdot 10^7$ a) the ^{236}U activity is slightly overestimated.

Table 6: Activities of Key Nuclides, Including Decayed Precursors, from Burning Th, U, Np and Pu (Bq per fission assuming a waste fraction of 1.0)

Nucl. burnt	Key nuclide	STS	PWR	FR	FMS	SFS
^{232}Th	^{233}U	9.95E-15	6.96E-15	1.50E-14	1.31E-14	9.95E-15
	^{234}U	4.09E-15	1.52E-15	3.00E-15	2.03E-15	9.08E-16
	^{231}Pa	3.72E-18	7.24E-17	5.42E-16	3.73E-15	6.43E-15
^{233}U	^{233}U	6.28E-14	8.45E-14	9.59E-14	1.04E-13	1.17E-13
	^{234}U	1.68E-14	1.75E-14	1.86E-14	1.58E-14	1.06E-14
^{235}U	^{236}U	4.20E-16	3.39E-16	3.29E-16	3.05E-16	2.38E-16
	^{237}Np	1.82E-16	1.07E-15	1.04E-15	8.94E-16	5.60E-16
	^{234}U	1.67E-16	1.04E-14	9.23E-15	6.12E-15	2.33E-15
^{238}U	^{236}U	3.61E-17	4.25E-18	5.03E-17	3.32E-17	8.90E-18
	^{237}Np	4.33E-17	3.42E-17	1.21E-16	1.24E-16	9.94E-17
	^{234}U	5.40E-19	1.12E-16	4.81E-16	5.98E-16	3.14E-16
	^{238}U	4.50E-18	4.83E-18	4.08E-18	4.17E-18	4.51E-18
	^{235}U	1.25E-18	2.47E-19	3.12E-18	3.05E-18	1.90E-18
^{237}Np	^{236}U	8.98E-17	6.23E-17	5.47E-17	2.99E-17	8.84E-18
	^{237}Np	6.29E-15	3.83E-15	4.33E-15	5.36E-15	7.28E-15
	^{234}U	6.42E-15	3.73E-14	3.82E-14	3.43E-14	2.30E-14
^{238}Pu	^{236}U	2.47E-16	9.37E-17	9.21E-17	5.87E-17	2.17E-17
	^{237}Np	2.96E-16	5.93E-16	2.30E-16	1.38E-16	4.59E-17
	^{234}U	2.31E-14	5.61E-14	6.46E-14	7.10E-14	7.96E-14
^{239}Pu	^{236}U	3.30E-16	1.99E-16	2.88E-16	2.31E-16	1.23E-16
	^{237}Np	3.96E-16	1.53E-15	6.37E-16	4.36E-16	1.47E-16
	^{234}U	4.68E-18	4.45E-15	2.24E-15	1.43E-15	4.62E-16
	^{235}U	6.56E-18	1.16E-17	1.79E-17	2.12E-17	2.65E-17
^{240}Pu	^{236}U	3.78E-16	2.74E-16	6.50E-16	7.01E-16	7.99E-16
	^{237}Np	4.54E-16	2.10E-15	1.43E-15	1.32E-15	9.49E-16
	^{234}U	5.33E-18	6.10E-15	5.00E-15	3.96E-15	1.87E-15
^{241}Pu	^{236}U	3.33E-16	2.06E-16	1.13E-16	6.95E-17	2.34E-17
	^{237}Np	4.91E-16	2.31E-15	4.10E-15	4.87E-15	6.32E-15
	^{234}U	5.76E-18	6.72E-15	1.41E-14	1.44E-14	1.24E-14
^{242}Pu	^{236}U	3.48E-16	2.72E-16	2.23E-16	1.58E-16	6.36E-17
	^{237}Np	6.96E-17	6.43E-16	3.92E-16	2.76E-16	1.09E-16
	^{234}U	3.88E-20	9.13E-16	1.00E-15	6.02E-16	1.74E-16
	^{238}U	2.44E-18	2.18E-18	2.81E-18	3.24E-18	3.98E-18
	^{235}U	3.26E-18	5.71E-18	4.45E-18	4.37E-18	3.47E-18

Table 7: Activities of Key Nuclides, Including Decayed Precursors, from Burning Am and Cm (Bq per fission assuming a waste fraction of 1.0)

Nucl. burnt	Key nuclide	STS	PWR	FR	FMS	SFS
^{241}Am	^{236}U	3.39E-16	1.37E-16	9.77E-17	5.85E-17	1.79E-17
	^{237}Np	4.90E-16	1.82E-15	3.53E-15	4.35E-15	6.01E-15
	^{234}U	1.38E-14	3.31E-14	2.85E-14	2.78E-14	2.29E-14
^{242m}Am	^{236}U	4.23E-16	3.34E-16	2.18E-16	1.68E-16	8.45E-17
	^{237}Np	8.99E-17	7.92E-16	3.89E-16	3.00E-16	1.46E-16
	^{234}U	1.00E-15	7.66E-15	4.13E-14	4.63E-14	5.49E-14
^{243}Am	^{236}U	4.28E-16	3.66E-16	4.86E-16	4.49E-16	3.37E-16
	^{237}Np	9.11E-17	8.66E-16	8.49E-16	7.30E-16	4.08E-16
	^{234}U	5.14E-20	1.23E-15	2.17E-15	1.55E-15	5.86E-16
	^{235}U	4.23E-18	7.68E-18	9.71E-18	1.24E-17	1.84E-17
^{243}Cm	^{236}U	4.14E-16	2.35E-16	3.62E-16	2.56E-16	7.51E-17
	^{237}Np	8.82E-17	6.49E-16	6.60E-16	4.32E-16	9.01E-17
	^{234}U	1.91E-19	1.12E-15	1.82E-15	1.10E-15	3.14E-16
	^{235}U	5.12E-18	1.56E-17	1.51E-17	2.05E-17	2.84E-17
^{244}Cm	^{236}U	4.89E-16	4.60E-16	6.86E-16	7.33E-16	8.11E-16
	^{237}Np	1.04E-16	1.09E-15	1.20E-15	1.19E-15	9.82E-16
	^{234}U	1.12E-20	1.54E-15	3.05E-15	2.39E-15	1.07E-15
^{245}Cm	^{236}U	6.47E-17	5.20E-17	1.53E-17	7.12E-18	3.55E-18
	^{237}Np	2.00E-16	1.59E-15	3.93E-15	5.89E-15	8.78E-15
	^{238}U	4.02E-18	3.37E-18	2.61E-18	1.89E-18	6.64E-19

An examination of the SFS/FR cumulative risk ratios in Table 9 and the fission–to–capture ratios in Table 1 does not reveal a simple relation between these ratios. It can be seen that, in contrast to the respective fission–to–capture ratios, the cumulative risks for the fissionable nuclides ^{237}Np, ^{238}Pu and ^{241}Am (i.e. nuclides which are of primary interest in connection with the burning of LWR–discharged minor actinides) show only modest spectrum hardening effects. As regards the fissile nuclides, ^{239}Pu shows a strong positive, and ^{241}Pu and ^{242m}Am show a negative spectrum hardening effect. This means that, in connection with risk considerations, the fission–to–capture ratio is not a useful parameter.

ACTINIDE BURNING IN HIGH–FLUX SUPERTHERMAL SYSTEMS

Table 8 indicates that high–flux superthermal systems can burn ^{237}Np slightly cleaner than other systems. This applies also for other important actinides with the exception of ^{232}Th, which burns cleaner in PWRs, and ^{239}Pu, which burns slighly cleaner in systems with a superfast neutron spectrum. The basic potential of high–flux superthermal systems for burning actinides is thus confirmed.

Table 8: Nuclide–Specific Risks from Burning ^{232}Th, ^{238}U, ^{237}Np and ^{239}Pu
(μSv per fission assuming a waste fraction of 1.0)

Nucl. burnt	Key nuclide	STS	PWR	FR	FMS	SFS
^{232}Th	^{233}U	5.37E-15	3.76E-15	8.08E-15	7.08E-15	5.37E-15
	^{234}U	1.31E-15	4.86E-16	9.59E-16	6.49E-16	2.91E-16
	^{231}Pa	1.56E-17	3.04E-16	2.28E-15	1.57E-14	2.70E-14
	Total	6.72E-15	4.57E-15	1.13E-14	2.34E-14	3.27E-14
^{238}U	^{236}U	1.08E-18	1.28E-19	1.51E-18	9.97E-19	2.67E-19
	^{237}Np	5.20E-17	4.10E-17	1.45E-16	1.48E-16	1.19E-16
	^{234}U	1.73E-19	3.58E-17	1.54E-16	1.91E-16	1.00E-16
	^{238}U	1.53E-18	1.64E-18	1.39E-18	1.42E-18	1.53E-18
	^{235}U	5.26E-18	1.04E-18	1.31E-17	1.28E-17	7.98E-18
	Total	6.00E-17	7.97E-17	3.15E-16	3.55E-16	2.30E-16
^{237}Np	^{237}Np	7.55E-15	4.60E-15	5.20E-15	6.43E-15	8.74E-15
	^{234}U	2.06E-15	1.19E-14	1.22E-14	1.10E-14	7.36E-15
	Total	9.62E-15	1.66E-14	1.74E-14	1.74E-14	1.61E-14
^{239}Pu	^{236}U	9.89E-18	5.98E-18	8.65E-18	6.92E-18	3.70E-18
	^{237}Np	4.75E-16	1.83E-15	7.64E-16	5.23E-16	1.76E-16
	^{234}U	1.50E-18	1.42E-15	7.17E-16	4.59E-16	1.48E-16
	^{235}U	2.76E-17	4.86E-17	7.52E-17	8.92E-17	1.11E-16
	Total	5.14E-16	3.31E-15	1.57E-15	1.08E-15	4.39E-16

Table 9: Ratios of Cumulative Risks from Burning Different Actinides

Nuclide	Type	FR/PWR	SFS/FR
^{237}Np	fissionable	1.05	0.93
^{238}Pu	fissionable	1.12	1.22
^{239}Pu	fissile	0.47	0.28
^{240}Pu	fissionable	0.74	0.53
^{241}Pu	fissile	1.91	1.22
^{242}Pu	fissionable	0.74	0.25
^{241}Am	fissionable	1.05	1.09
^{242m}Am	fissile	3.98	1.30
^{243}Am	fissionable	1.20	0.43

On the other hand, due to their poor overall neutron balance, high–flux superthermal systems will not be able to play a dominant role as energy producers in a long–term strategy. They are suitable for burning existing actinide wastes which cannot be recycled in other systems, and remaining actinide wastes from closed conventional systems.

THE URANIUM–PLUTONIUM AND THE THORIUM–URANIUM FUEL CYCLE

The ultimate goal in nuclear energy is to extract the fission energy from all ^{238}U and ^{232}Th with a minimum long–term risk for the population. The numbers in Table 2 indicate that, from the viewpoint of the overall neutron balance, this goal can be achieved best with uranium and fast reactors. For thorium, the surplus of neutrons may not be sufficient to compensate for neutron losses. Therefore, the benefits from incorporating accelerators into the systems may be more significant for thorium–based than for uranium–based systems.

As to the goal of a small long–term risk for the population, Table 8 indicates that, in spite of the reduced build–up of higher actinides, thorium does not perform better than uranium: For ^{232}Th, the risk is dominated by ^{233}U and – in systems with a very fast neutron spectrum – by ^{231}Pa. ^{237}Np and ^{234}U contribute about equally to the risk from burning ^{238}U. It can be seen that, for all systems and even without the ^{231}Pa contribution (^{231}Pa could be separated and burnt in a thermal system), the comparison favours the uranium–plutonium fuel cycle.

SUMMARY AND CONCLUSIONS

Based on the concepts of the asymptotic actinide inventory and the key nuclides for radiological risk assessment, a general method for investigating the effectiveness of actinide transmutation systems has been developed. The method allows to assess the impact of different long–term transmutation strategies on the actinide inventories of the systems, the composition of the actinide waste, and the radiological risk associated with the disposal of this waste. In a comparative study, the method was applied to systems with a wide range of characteristics including a PWR, a fast reactor, a high–flux superthermal system, and two hybrid systems with fast neutron spectra. The principal results of the study can be summarised as follows:

- A complete burning of both fissile and fissionable actinides requires an adequate overall neutron balance. In general, this can only be achieved in systems with a fast neutron spectrum.
- The neutron spectrum can be hardened and the neutron balance can be improved by coupling an accelerator with the system. This may be particularly interesting for thorium–based systems which are less abundant in neutrons than fast systems with uranium–based fuels.
- For comparable fuel burnup and reprocessing losses, the recycling of the important transuranic actinides in fast reactors and PWRs results in similar radiological risks of the waste. Risk ratios of more than a factor of two arise in two cases: ^{239}Pu burns cleaner in a fast, and ^{242m}Am burns significantly cleaner in a thermal neutron spectrum.
- It appears that the radiological risk of the waste is not directly related to the mean neutron energy of the system or the type of nuclide being burnt. Burning fissionable nuclides in a thermal neutron spectrum is possible from the viewpoint of the risk, but undesirable, because it necessitates the use of additional fuel which indirectly augments the risk, or a hybrid system.
- With the exception of ^{232}Th and ^{239}Pu, actinides burn with the smallest risk in high–flux superthermal systems. In addition, these systems feature low actinide inventories and, consequently, also small core accident risks.
- The present analysis indicates that, in the long range, the radiological risk of the waste cannot be reduced by changing from the uranium–plutonium to the thorium–uranium fuel cycle.

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