C H A P T E R I

THE FUEL CYCLE OPTIONS AND THE RADIOLOGICAL SOURCE TERMS

INTRODUCTION - SCOPE

The purpose and scope of this document is to survey the availability and quality of data on which to base a systems study of partitioning and transmutation (P&T) as an alternative waste management option.

The global approach on nuclear power development will underly this assessment study and emphasise the merits and limitations of the P&T option on the world wide environmental and radiological impact of the nuclear electricity production.

The concept of partitioning goes far back into the history of nuclear development and as early as the mid-sixties important R&D programmed were launched to investigate the separation technology of long lived fission products (^{137}Cs, ^{90}Sr) from High Level Liquid Waste (HLLW). Soon it was recognised that any global improvement of the HLW impact required an integrated approach incorporating the minor actinides in order to be relevant for the long term.

In the early seventies however, the permanent presence of very toxic radionuclides for thousands of years in geologic repositories was deemed a risk, which ought to be avoided. Alternative scenarios comprising the separation of minor actinides from HLLW and transmutation of these nuclides in LWR, FBR and HTGR received considerable attention until it was recognised that geological disposal of radionuclides cannot be avoided and that all P&T scenarios end up with an actinide concentrate or residue which has to be stored and eventually disposed of.

Extensive information on all aspects of P&T was published in the proceedings of the first and second technical meeting on the Nuclear Transmutation of Actinides held in 1977 and 1980. [1] [2].

These conferences were followed by a period of extensive assessment of data which were published by JRC in 1983 and 1984 [3] [4].

A parallel investigation programme was undertaken in the US and led to the conclusion that the radiological impact of long-lived fission products (^{129}I, ^{99}Tc) on the biosphere is equally or even more important than that of the minor actinides.

A particular effort was undertaken by the "Conseil Supérieure de la Sureté Nucléaire" in France which published the well known CASTAING report in 1982 [6]. These matters were very recent 1 y re-exami ned by a group of experts of the French CEA [7] and the P&T issue received a new political back-ing.

In 1988 the Japanese Government launched a very ambitious long term programme, called OMEGA, which covers the following items [8] [9] [10] :

recovery and possible utilisation of fission products, minor actinides and noble metals in the frame work of an advanced reprocessing option;

transmutation of long lived actinides and fission products by LWR, FBR and Accelerators.

NUCLEAR FUEL CYCLE OPTIONS

presently there are two main options in the back end of the fuel cycle :
the once through cycle;
the conventional reprocessing cycle with U-PU recycling.

The advanced reprocessing cycle with additional P & T operations on waste from conventional reprocessing requires additional facilities which have to be attached to the existing reprocessing plants.
By reviewing the Nuclear Power Plant programme (1990) a subdivision can be made as follows :

	GWe	%
- OECD Countries committed to Reprocessing and associated partners (FR, UK, JAP + clients)	134,4	= 40
- NON OECD Countries principally committed to reprocessing (USSR + Associates)	47,-	14
- OECD Countries committed to direct disposal (USA, SP, SW, CAN)	137,3	= 41
- Countries outside OECD	18,4	≃ 5
	337,-	100

Table I gives in more detail the respective NPP programmed per country and the expected evolution of their reactor parks. As a whole we may subdivide the world into two almost equal blocks of 54 and 46% adhering to a different fuel cycle option.

Beyond the year 2000 only estimations can be made based on electricity growth scenarios. A recent OECD evaluation for a low growth scenario led to the following data (GWe) :

	2010	2020	2030
OECD North America	148	174	196
OECD Europe + Japan	229	284	348

The other countries do not markedly influence the projections.

Advanced reprocessing is only conceivable in those countries where a reprocessing activity already exists. The USA occupies a dual position as a non reprocessing country with a potential capacity and know how in reprocessing facilities.

A second factor which must be considered is the reprocessing capacity and the access to sensitive nuclear technology.
Table II gives an overview of the existing or planned reprocessing facilities for civilian purposes.

TABLE I I

Nominal reprocessing capacities in THM/year of existing plants and of units under construction

	Existing Nominal capacity	Capacity Under construction
La Hague	1200	1600
Marcoule (GG)	600	
Sellafield - (MAGNOX) - (THORP)	1200	- 1200
Tokai	200	200
Rokkashomura		800
SAVANNAH RIVER	p.m.	

A number of countries outside OECD, except USSR and CHINA will have very difficult access to sensitive fuel cycle technology and will as a consequence be forced to take the non reprocessing option or to sign reprocessing agreements.

Unless the military facilities would be reconverted to civilian reprocessing or to advanced reprocessing facilities there will be no major change in the general picture on the reprocessing issue before the year 2000.

In order to link the nuclear electricity production to the amount of spent fuel to be stored or to be reprocessed the NPP electricity output has to be multiplied by the discharged fuel mass. 1 GWe capacity at a 0.75 load factor and at a burnup of 33,000 MWd/T would discharge about 27 THM. With increasing burnup the spent fuel out-put will be reduced to about 23 THM/GWe year at 40,000 MWd/T. The tendency being to increase burnup in order to reduce fuel costs, it is reasonable for the future to take a mean value of 25 THM/GWe.

By multiplying the data of Table I with a range of values for spent fuel one obtains the total amount of spent fuel discharged yearly. This quantity has to be compared to the reprocessing capacity existing at the present time and the capacity which would be required to treat the total inventory.

TABLE I

Overview of the Nuclear Power Plant programmed and subdivided according to their fuel cycle options.
[present and future evolution]

	Operational	In Construction	Planned
Countries committed to REPROCESSING			
OECD countries			
FRANCE	54.746	12.762	7.448
U.K.	15.102	1.258	3.750
JAPAN	29.445	12.546	14.484
GERMANY FR	23.927	327	-
OR	1.835	2.640	(1.760)
BELGIUM	5.749	-	
SWITZERLAND	3.065	1.214	
NETHERLANDS	539		
SUBTOTAL	134.408	30.747	27.442
EAST EUROPEAN COUNTRIES.			
ex USSR	36.636	25.000	34.980
BULGARIA	2.760	3.000	2.000
CZECHOSLOVAKIA	3.434	5.784	6.084
FINLAND	2 .400	1.000	
HUNGARY	1.760	5.000	-
POLAND		1.860	-
ROMANIA		3*395	-
SUBTOTAL	46.990	45 .039	43.064
Countries not Committed to Reprocessing			
OECD Countries			
USA	106.773	4.610	-
SPAIN	7.852	-	-
SWEDEN	10.130	-	-
CANADA	12.603	3.740	-
SUBTOTAL	137.358	8.350	-
OTHER Countries			
SUBTOTAL	18.401	9.890	25.157

Table III shows the data for the main group of countries involved in nuclear power.

TABLE III

Spent fuel arisings per year compared to existing reprocessing capacity and required increment to process all spent fuel (in THM/y)

Power capacity GWe	Spent fuel* arisings in	Reprocessing capacity	Needed increment of Reprocessing capacity
OECD-REPRO 134	3.080 - 3.620	3.800	
non OECD-REPRO 47	- 1.270	(1.000)**	270
OECD NON REPRO 137	3.150 - 3.700		3.150 - 3.700
Others NON REPRO 18	414 - 486		414 - 486
Totals	7.724 9.076	4.800	3.564 - 4.456

* Minimum 23 T/GWey - Maximum 27 T/GWey

** Planned capacity in the USSR

In order to support the advanced reprocessing option the present reprocessing plants operating or in construction should be adapted to include the partitioning flowsheets for minor actinides and long lived fission products. In the countries not committed to civilian reprocessing an additional processing capacity of 3500 to 4500 THM/year has to be constructed.

Since only a limited reprocessing capacity has been operating in the past decades within the OECD countries, a spent fuel back log of 84.000 THM has piled up and this inventory is expected to grow till 200.000 THM in the year 2000.

Potentially the advanced reprocessing option might be taken by the OECD reprocessing minded countries as their annual processing capacity will approach the spent fuel arisings per year. This option will undoubtedly decrease the radiological impact of the national or regional waste repositories.

From global point of view such a decision would not reduce worldwide the radiological impact significantly since only 53% of the total spent fuel arisings are involved.
A number of countries representing at present only about 5% of the total spent fuel output, will in any case not be able to make an alternative choice to direct disposal.

A further glance on table I shows, that, if all in-construction and planned facilities are realized, a total world capacity of 525 GWe would become available. The reprocessing minded countries make up a total 62% and the non-reprocessing option is then reduced to 38% among which 10% result from developing nations without access to reprocessing technology.

If advanced reprocessing is implemented in an ideal fashion on the majour portion of the spent fuel a maximum reduction with a factor of 20 may be expected on the radiological impact of the very long lived radionuclides (^{237}Np, ^{129}I ...).
This global limitation has to be kept in mind when defining technical goals of advanced reprocessing for certain radionuclides.

As a conclusion we might say that in the most optimistic case between 90 and 95% of the spent fuel arisings might be reprocessed.
The maximum hypothesis for advanced reprocessing is to consider that al 1 spent fuel resulting from nuclear power plants by the year 2030 will be reprocessed and recycled through Fast Reactors. This is a pure working hypothesis which will be used for estimating the future needs of P & T facilities.

RADIOLOGICAL AND CHEMICAL SOURCE TERM OF SPENT FUEL AND HLW

The conventional source term of radionuclides is based generally on the ORIGEN (2) and KORIGEN calculated compositions of spent fuel [11].[12]. The data are given for PWR-UO_2 spent fuel in table IV and for PWR-MOX spent fuel in table V in each case for a standard burnup of 33.000 MWd/T, 5 years after discharge from the reactor.

For reasons which will be explained below, only half lives exceeding about 30 years (e.g. ^{90}Sr and 137 Cs) have been taken into consideration except for 241 Pu because of its direct relation with 241 Am which is of major importance for further discussion in this report.

The data for HLW compositions have been considered identical to those of spent fuel except for U and Pu isotopes which are supposed to be extracted for 99 % (a mean value between 100 and 98 %). Iodine 129, which is separated during reprocessing and transferred to MLW and/or discharged into the sea, is taken into consideration in this study because of its radiological importance associated with its very long half life and its biological concentration.

Data for higher burnups (up to 50.000 MWd/T) have not been extensively documented since there is a good proportionality between burnup and fission product content on the one hand and only slight modifications in the actinide concentration-distribution.

The radioactive source term can be derived from the activity in Ci/THM (Tables IV and V) multiplied by the yearly spent fuel output (Table III) from the nuclear electricity programmed.

In the countries with the non-reprocessing option every THM charged into the reactor, leaves it as a waste material (3560 - 4450 THM/y), U and Pu included. The reprocessing option produces worldwide a segregated U stream of 4160 to 4890 THM/y and a Pu source term of 38.6 - 45.4 T per year which are in principle intended to be reused. An annual residual amount of 48 T U and 0.45 T Pu together with the fission products is declared waste. (HLW)

Among the major actinides (U and Pu) a factor of 100 exists between the reprocessing and the non reprocessing option. However the total environmental impact of the fuel cycle with reprocessing is not 100 times smaller since the separated U has to be stored as UF_6, UF_4 or U_3O_8 and as the Pu reuse is limited to one or two recycling in LWR's no overall Pu reduction is observed between the charged and the discharged fuel.

The radiotoxicity of the spent MOX fuel is even slightly higher than that of the initial MOX load because of the higher residual Pu content and an increased MA concentration. The overall Pu balance in MOX fuel led reactors results from a Pu generation in the UO_2 fuel elements and a depletion in the MOX fuel elements.

But even taking into account these inherent limitations of LWR recycling, the overall environmental impact reduction is significant for the reprocessing option.

However, in the medium term, a solution must be found for the spent MOX fuel elements or for the stored Pu.

TABLE IV

Actinides and Long Lived Radionuclides composition of PWR - UO_2 spent fuel. and its HLW equivalent at 33.000 MWd/T after 5 years cooling.

ACTINIDES				
NUCLIDE	t1/2(y)	Ci/THM	g/THM	HLW FRACTION
U 232	70	$3.27\ 10^{-2}$	-	
U 234	$2.44\ 10^{5}$	0.94	150	
U 235	$7.\ 10^{8}$	$1.79\ 10^{-2}$	8250	1% to
U 236	$2.34\ 10^{7}$	0.262	4050	HLW
U 238	$4047\ 10^{9}$	0.317	943000	
Np 237	2.14 10	0.308	437	100% to HLW
Pu 238	87.7	$2.39\ 10^{3}$	140	
Pu 239	24110	340	5470	
Pu 240	6550	509	2230	1% to
Pu 241	14.4	$9.85\ 10^{4}$	956	HLW
Pu 242	$3.7\ 10^{5}$	1.86	486	
Am 241	432.6	1002	296	
Am 242	142	3.8	0039	
Am 243	7370	16.7	83.8	100% to HLW
Cm 243	28.5	14.8	-	
Cm 245	8500	0.138	0.804	
Cm 246	4730	0.225	0.731	

FISSION PRODUCTS				
NUCLIDE	t1/2(y)	Ci/THM	g/THM	HLW FRACTION
Sr 90	28.5	$6.63\ 10^{4}$	485	
Y 90	equil 6	$6.63\ 10^{4}$	0.126	
Zr 93	$1.5\ 10_{5}$	1.79	736	
Tc 99	2.1 106	13.2	841	100 % to
Pd 107	6.5 10	0.102	231	HLW
Sn 121	50_{5}	0.146	4.5	
Sn 126	10^{5}	0.542	19.2	
I 129	$1.57_{6}10^{7}$	$3.12\ 10^{-2}$	229	0.1% to HLW
Cs 135	$2\ 10^{6}$	0.352	324	
Cs 137	30.1	$9.5\ 10^{4}$	1100	100 % to
Ba 137	equil.	$9.4\ 10^{4}$	178	HLW
Sm 151	93	324	44.2	
LIGHT ELEMENTS				
c 14	5730	0.143	0.0315	0.1 % to HLW
Ni 59	$7.5\ 10^{4}$	17.5	216.6	100% to
Ni 63	100	24.10^{3}	24.28	HLW

TABLE V

Actinides and Long Lived Radionuclides composition of PWR - MOX spent fuel.
at 33.000 MWd/T after 5 years cooling.

ACTINIDES					FISSION PRODUCTS				
NUCLIDE	t1/2(y)	Ci/THM	g/THM	HLW FRACTION	NUCLIDE	t1/2(y)	Ci/THM	g/THM	HLW FRACTION
U 232	70	9.4110-3	1.39 10-4		Sr 90	28.5	3.14 10^4	230.3	
U 234	2.44 10^5	0.321	51.44		Y 90	equil.	3.14 10^4	0.057	
U 235	7. 10^7	2.36 10^{-3}	1093	1% to	Zr 93	1.5 10^6	1.24	494.4	
U 236	2.34 10^7	1.4 10^{-2}	216.9	HLW	Tc 99	2.1 10^5	13	766	100 % to
U 238	4.47 10^9	0.311	925900		Pd 107	6.5 10^6	0.266	517.6	HLW
Np 237	2.14 10^6	9.02 10^{-2}	128.3	100% to HLW	Sn 121	50	00341	5.76 10^{-3}	
Pu 238	87.7	1.64 10^4	960.6		Sn 126	10^5	1.04	36.6	
Pu 239	24110	863.7	13890		I 129	1.57 10^7	0.0413	234	).1% to HLW
Pu 240	6550	2770	12150	1% to	Cs 135	2 10^6	0.667	579.3	
Pu 241	14.4	5.33 10^5	5177	HLW	Cs 137	30.1	9.3 10^4	1069	.00 % to
Pu 242	3.7 10^5	13.32	3487		Ba 137	equil.	8.8 10^4	170.5	HLW
Am 241	432.6	6705	1953		Sm 151	93	765	20	
Am 242	142	158.1	16.34						
Am 243	7370	214.1	1074	100%	LIGHT ELEMENTS				
Cm 243	28.5	313.7	6.07	to HLW					
Cm 245	8500	5.27	30.71		C“14	5730	1.13 10-4	2.55 10^{-5}	0.1 % to HLW
Cm 246	4730	0.82	2.66		Ni 59	-			100% to
					Ni 63	-			HLW

This is the case in an advanced fuel cycle scheme with Fast Reactors, where the effect of the U and Pu reuse can further be optimised and could approach a factor of 100 in radiological impact between the once through cycle and the reprocessing option if a closed cycle of U and Pu is realised. However presently we are very far from a worldwide industrial closure of the fuel cycle and the P & T option of minor actinides and long lived fission products has to be assessed as a working hypothesis.

Table VI shows the actinide concentration in HLW as a function of long term cooling time. It is obvious that decay does not improve the situation very much after 1,000 years which is the protective barrier life time in a geological repository structure as defined by the NRC in 10 CFR 60.

Important decay is observed for Pu 238 and Pu 241 while the minor actinides remain roughly at the same level or do show an increase as is the case for Np 237.

Table VII shows similar data for fission products. It is striking that only a few fission products influence the long term radiological impact a.o. Zr 93, Tc 99, Sn 126, I 129 and Cs 135.

The major fission products Sr 90 and Cs 137 play only a role during the first 500 years which is within the lifetime of the engineered barriers. Some useful applications of Cs 137 as substitute for Co 60 might also be considered in the technico-economic evaluation of P & T.

The fission product fraction contains a non negligeable noble metals content which is from ressource preservation point of view an incentive for the advanced reprocessing option with an additional P & T step.

The source terms resulting from the three different fuel cycle options can be summarised as follows (quantities per THM) :

TYPE OF NUCLIDES	NON-REPROCESSING (SPENT FUEL)	CONVENTIONAL REPROCESSING (HLW)	ADVANCED REPROCESSING (P&T)
Major actinides	955kg U+9.3kgPu	10kg U + 93gPu	10kg U+9.3gPu
Minor actinides	0081 kg	0.81 kg	0.081-0.0081kg
Fission products	35 kg	35 kg	30kg + 5 kg reusable

TABLE VI

Long term decayof actinides resulting from PWR-UO_2 irradiated at 33.000 MWd/T 1 % U+Pu and 100% Minor Actinides in HLW.

HLW from Reprocessing 7 years after discharge

	g/THM				Ci/THM			
Decay time " (y)	50 Y	100 Y	1000 Y	10.000 Y	50 Y	100 Y	1000 Y	10.000 Y
234	1.92	2.26	3.17	3.11	1.2 10^{-2}	1.41 10^{-2}	1.98 10^{-2}	1.95 10^{-2}
U 235	82.6	82.7	84.2	103.0	1.79 10^{-2}	1.79 10^{-3}	1.82 10^{-3}	2.23 10^{-3}
U 236	40.6	40.8	44.4	66.4	2.63 10^{-3}	2.64 10^{-3}	2.87 10^{-3}	4.3 10^{-3}
U 238	9430	9430	9430	9430	3.17 10-^{3}	3.17 10^{-3}	3.17 10^{-3}	3.17 10^{-3}
Np 237	463	491	744	820	0.327	0.346	0.524	0.578
Pu 238	1.03	0.735	5.2 10^{-3}	-	17.6	12.6	8.6 10-^{2}	-
Pu 239	5501	55.5	60.7	83.9	3.43	3.45	3.77	5.12
Pu 240	36.8	39.5	36.4	14	8.38	9.03	8.3	3.2
Pu 241	1.1	0.1	1.24 10^{-3}	5.94 10-^{4}	113	10.3	0.128	6.12 10^{-2}
Pu 242	4.88	4.89	5.01	5.38	1.86 10-^{2}	1.87 10^{-2}	1.92 10^{-2}	2.06 10^{-2}
Am 241	364	337	79.7	1.79 10-^{2}	1250	1160	273	6.14 10^{-2}
Am 242 m	0.317	0.253	4.1810-3	-	3.09	2.46	4.06 10^{-2}	-
Am 243	83.5	83.1	76.3	32.8	16.6	16.6	15.2	6.54
Cm 243	9.61 10^{-2}	2.85 10^{-2}		-	4.96	1.47	-	-
Cm 245	0.80	0.798	0.741	0.356	0.138	0.137	0.127	0.061
Cm 246	0.726	0.721	0.632	0.169	0.223	0.222	0.194	0.052

TABLE VII

Long term decay of fission products and noble metals inventory of spent fuel from PWR-UO_2 at 33.000 MWd/T (100 % to HLW).

NUCLIDE	FISSION PRODUCTS						
	Ci/THM				STABLE NUCLIDE	g/THM	
	5oy	looy	1000y	looooy			
Sr 90	$2.27\ 10^{4}$	6920			Zr 90	485	
Y 90	$2.27\ 10^{4}$	6920	-	-	Zr 90	0.126	
Zr 93	1.79	1.79	1.79	1.79	Nb 93	736	
Tc 99	13.2	13.2	13.1	12.8	Ru 99	841	
Sn 107	$7.8\ 10^{-2}$	3*91 10-^{2}	-	-	Sb 121	4.5	
Sn 126	0.542	0.542	0.538	0506	Te 126	19.2	
I 129	$3.12\ 10^{-2}$	3.1210-2	$3.12\ 10^{-2}$	$3.12\ 10^{-2}$	Xe 129	229	0.1% to HLW, 99.9 to MLW
Cs 135	0.352	0.352	0.352	0.351	Ba 135	324	
Cs 137	$3.36\ 10^{4}$	$1.06\ 10^{4}$	-	-	Ba 137	1100	
Ba 137	$3.18\ 10^{2}$	$1.0\ 10^{2}$	-	-	Ba 137	178	
Sm 151	$2.29\ 10^{2}$	$1.56\ 10^{2}$	-	-	Eu 151	44.2	
NOBLE METALS							
Ru element	-				Ru	2106	Ru 99,100,101,102,103,104,105,106
Rh element	-				Rh	4.14	Rh 102,103,106
Pd element					Pd	1258	Pd 104,105,106,108,110
Pd 107	0.102	0.102	0*102	0.102	Ag107	(231)	(included in 1258 g/T Pd)

CUT-OFF HALF LIFE FOR P & T OPERATIONS.

The cut-off half life is the time interval below which it has no sense to carry out transmutation in order to produce nuclides with shorter half lives.

In order to set realistic objectives to minor actinides and long lived fission products P & T operations, we must investigate whether and to what extent,' any supplementary treatment is counter-balanced by real radiological-. and waste management benefits. Some economically exploitable resources e.g. Cs 137 as a radiation source in food irradiation technology and Ru-Rh-Pd concentrates as additional noble metals source could be of some help in improving the economic viability of the P & T option.

A P & T process must ultimately aim at a realistic half life reduction from thousands of years to some decades. The lower limit of this cut off is a trade-of between treatment cost and radiological benefit. Any P & T operation which aims at reducing the half-lives of the separated nuclides below 25 to 30 years runs up against the massive presence of Sr 90 and Cs 137 in HLLW.

Partitioning of Cs 137 is technically possible, though much more difficult for Sr90, but transmutation of these nuclides seems according to the present state of the art very speculative or expensive.

The present practice of prolonged intermediate storage of spent fuel before reprocessing also contributes to the elimination of short lived nuclides.

The tendency to increase the fuel burnup will complicate conventional reprocessing by producing more secondary waste and increase the collective dose to operating personnel. These negative elements can also be neutralised by prolonged cooling before reprocessing.

However this cooling time cannot be lengthened indefinitely without jeopardizing the enormous investments done in the' large reprocessing plants.

In the course of this study a cut-off period of 30 years will be used in the assessment of the P & T influence on the management of waste streams from fuel cycles.

RADIOLOGICAL HAZARD RANKING OF MINOR ACTINIDES AND LONG LIVED FISSION PRODUCTS.

The general strategy of introducing P & T as an alternative waste management option is based on the radiological benefit which is expected from such a venture. The selection of the actinides and long lived fission products which are beneficial to eliminate by transmutation can be made on the comparison of their hazard factors.

The notion of hazard criterion was introduced by CLAIBORNE in 1975 [14] and relies on a very simple concept i.e. the volume of drinking water required to dilute a mixture of radionuclides to the drinking water limits. These data are isotope specific and actualised according to the present drinking water limits.

The drinking water and groundwater limits incorporated in the present legislative documents [15] are based on the ICRP guidelines of 1980. But recently ICRP issued a new set of guidelines for radionuclides in publication 61 [17]. These data are generally more stringent for Actinides except Np 237 whose ALI has been increased by a factor of 10. With a few exceptions the same trend is found for the long lived fission products whose ALI's have been generally decreased with a factor of 3.

The drinking water limits and associated hazard factors are given in Table VIII for actinides and in Table IX for long lived fission products.

The technical lifetime of an engineered barrier ranges from 200 to 1000 years. In order to select those radionuclides which are prime candidates for P & T we have to consider the hazard factors and the technical possibility to isolate the radionuclide from the others in order to prepare them for transmutation.

In terms of hazard factors the following listing can be made for actinides in HLW.

ACTINIDES HAZARD FACTOR RANKING IN HLW

	ICRP 30	l /THM		ICRP 61	l /THM
1	**Am 241**	2 -7 10^{13}	1	Am 241	3.3-12.31013
2	Am 243	1.1 10^{12}	2	Am 243	1.8 -2 10^{12}
3	**Np** 237	4.7-6.41011	3 (l%)	pu 240	7.6-8 10^{11}
4 (l%)	**Pu** 240	1.5 10^{11}	4 (l%)	**Pu** 239	3.2-3.51011
5 (l%)	**Pu** 239	6.4-6.91010	5	Np 237	4.7-6.41010
6	Cm 246	1.4-1.61010	6	Cm 246	2.4-2.71010

The ranking list relies on a 100 % transfer of minor actinides from the fuel to the HLW and on 1 % of U and Pu in the residues. The first two radionuclides, Am 241 and Am 243 are by far the most important ones from radiologic point of view, whether the ICRP 30 or 61 values are taken into account.

A change of order appears at the 3rd to 5th positions. The recent tenfold increase of the Np 237 ALI value coupled to a decrease of the Pu 239-240 ALI's with a factor of 5 is at the origin of this shift in ranking. Even hypothesizing that the Pu decontamination from HLW can be reduced in the future e.g. with a factor 5 or 10, the new ranking will not be modified. However the previous discussion shows how variable radiotoxicological data are and how they depend on advances in radiobiological research and on its interpretation.

Neptunium which was considered a prime radiological hazard by ICRP 30 is only marginal compared to Am and Pu if ICRP 61 is taken into account. However in the very long run (hundred-thousands or million years) Neptunium 237 is still a very important hazardous actinide.

The possibility to isolate a nuclide from HLW depends on the nature of the nuclide involved and the mass of chemically similar elements which behave in the same way as the nuclide to be separated e.g. the rare earths. It is obvious that any separation procedure of an actinide must be preceded by chemical reprocessing which is the indispensable first step in any P & T operation.

If P & T is performed on HLLW the possibility exists to reduce the radiological impact of the subsequently vitrified waste by a significant factor.

The ranking of the hazard factors of spent fuel in the 200 - 1000 year period is according to ICRP 30 and 61 as follows :

HAZARD FACTORS OF SPENT FUEL

	CRP 30 nucl ide	1 /THM		CRP 60 nucl ide	1 /THM
1	Am 241	2 -7 10^{13}	1	Al 241	3.33-11.6 10^{13}
2	pu 240	1.5 -1.6 10^{13}	2	pu 240	7.5 - 8 10^{13}
3	pu 239	6.45-6.91012	3	pu 239	3.2-3.451013
4	Am 243	1.1-1.21012	4	Am 243	1.8 - 2 10^{12}
5	Np 237	4.7-6.41011	5	Pu 242	1.72 10^{11}
6	pu 242	1.3 10^{10}	6	Np 237	4.7 - 6.4 10^{10}
7	Cm 246	1.4-1.61010	7	Cm 246	2.3 - 2.661010

Americium 241 remains at the top with a hazard coefficient which isonly slightly different from that of HLW. Between 1000 and 10.000 years Am 241 decays gradually to negligible levels and loses its predominant radiotoxic position to Pu 240-239.

By comparing the hazard ranking list of HLW and spent fuel it appears that conventional reprocessing does not decrease the radiotoxicity within the first 1000 years, but it is a necessary step to reduce this long term toxicity. The only alternative to P&T would be a pure transmutation scenario by which spent fuel would quantitatively be transformed into fission products or short lived actinides without a foregoing partitioning step. However such a scenario is very speculative. Advanced reprocessing with P&T incorporated in the process is from pure radiological point of view preferable to spent fuel storage and conventional reprocessing on the condition that the transmutation step results in a quantitative elimination of the separated long lived actinides. The long lived fission products are much less toxic than actinides on the basis of hazard indexes once Sr 90 and Cs 137 have decayed i.e. after about 600 y. In order of significance, the following radiotoxicity ranking can be derived from the data of Table IX in the 200 - 1000 y period on the basis of ICRP 61.

RANKING OF FISSION PRODUCTS

		Nuclide	1 /THM
	1	Sr 90	3.91012 -2.13 10^{4}
	2	Cs 137	3.81012 - 3.66 10^{4}
(1)	3	Tc 99	1.6 10^{9}
(2)	4	Sn 126	6.6 10^{8}
(3)	5	I 129	5.8 10^{8}
(4)	6	Cs 135	1.3 10^{8}
(5)	7	Zr 93	9.4 10^{7}

Abstraction made of Cs 137 and Sr 90 which decay within 600 years, I 129 and Tc 99 are very important nuclides because of their long half lives and mobility in the geosphere.

Iodine 129 deserves a special discussion in the context of ranking the nuclides because of its half life of 1.57 10^{7} years and its chemical properties which do not lend this element to a stable matrix. The combination of both factors brings about that any confinement technology whatsoever

cannot guarantee a separation from the biosphere. Morever iodine is a very mobile element in the geosphere and migrates much more rapidly than e.g. Pu, Am... Calculations in the frame work of the PACOMA project of the CEC [16] have shown that in a normal scenario the I 129 activity would reach the neighboring aquifers after about 5000 years. The radiologic influence of that long term radionuclide is such that it represents a dose contribution of 10 % of the natural background.

Tc 99 is a fission product with a half life of 2.1 10^5 years which is mobile in some strata e.g. tuff and-contributes largely to the dose to man in oxidative aquifers. In reducing conditions (clay) it is not mobile.

During the reprocessing steps it behaves partly as a platinum type metal, partly as a higher oxide. Although the hazard ranking factor is 10^5 times lower than that of actinides its contribution to the dose to man is very important.

The other fission products Sn 126, Cs 135, Zr 93 are in decreasing order much less important in the radiological context.

EFFECT OF THE GEOLOGIC CONFINEMENT ON HAZARD RANKING

In the actually accepted fuel cycle options (spent fuel storage or vitrified HLW disposal) the geologic confinement within a multiple barrier system in a well chosen host rock, plays a dominant ro"le in the assessment of the ultimate radiological safety of the nuclear fuel cycle.

The integrity of a suitable geologic disposal system depends on the intrinsic confinement of a host rock which is supposed to be very impermeable to water (e.g. clay, impermeable granite) or untouched by surface waters (e.g. salt layers or domes) during geological periods.

In these favorable cases, the confinement extends over hundred thousands to million of years and influences the hazard ranking of actinides.

After 5000 years, Am 241 loses its first ranking hazard factor to Am 243 and Np 237. However in the very long term, Np 237 is the critical nuclide because it has a half life (2.14 M years) which surpasses the geologic integrity interval of most geologic layers.

The ranking of the fission products is not altered by the role of a geological disposal option. Tc 99 and Iodine 129 with a half live of 2.1 10^5 and 15.7 M years respectively remains high on the list of the fission products to be examined in a P & T scenario.

TABLE VIII

PERMISSIBLE CONCENTRATIONS AND HAZARD FACTORS OF ACTINIDES IN HLW EXPRESSED PER THM.

Nuc 1 ide	Ground & Drinking water limits Bq/l (*)		HAZARD FACTORS 1/THM						Remark
			5 y	after 200 y	1000 y	5y	after 200 y	1000 y	
	(1980) (ICRP 30)	(1990) (ICRP 61)	according ICRP 30			according ICRP 61			
U 232	0.8	2	1.51 10^{7}	2.08 10^{6}	7.58	6. 10^{6}	8.3 10^{5}	3.03	
U 234	4	7	8.67 10^{7}	1.55 10^{8}	1.83 10^{8}	4.9 10^{7}	8.8 10^{7}	1.04 10^{8}	to be multiplied by 10^{2} for spent fuel
U 235	5	7	1.32 10^{7}	1.32 10^{6}	1.32 10^{5}	9.4 10^{5}	9.4 10^{5}	9.4 10^{4}	
U 236	5	7	1.94 10^{7}	1.97 10^{7}	2.12 10	1.38 10^{7}	1.38 10^{7}	1.51 10^{7}	
U 238	5	8	2.34 10	2.34 10	2.34 10^{7}	1.46 10	1.46 10	1.46 10	
Np 237	0.03	0.3	3.761011	4.7 10^{11}	6.43 10^{11}	3.761010	4.7 10^{10}	6.431010	
Pu 238	3	0.4	2.94 10^{11}	8.1 10^{10}	1.06 10^{9}	2.2 10^{12}	6.07 10^{11}	7.95 10^{9}	to be multiplied by 10^{2} for spent fuel
Pu 239	2	0.4	6.3 10^{10}	6.45 10^{10}	6.951010	3.15 10^{11}	3.2 10^{11}	3.47 10^{11}	
Pu 240	2	0.4	9.4 10^{10}	1.6 10^{11}	1.53 10^{11}	4.7 10^{11}	8 10^{11}	7.65 10^{11}	
Pu 241	100	20	3.641011	8.1 10^{7}	4.73 10^{8}	1.8 10^{12}	4 10^{8}	1.89 10^{8}	
Pu 242	3	0.4	2.3 10^{8}	2.31 10^{9}	2.36 10	1.7 10^{9}	1.7 10^{9}	1.77 10	
Am 241	0.5	0.3	7.4 10^{13}	7.2 10^{13}	2 10^{13}	1.23 10^{14}	1.2 10^{14}	3.33 10^{13}	
Am 242	0.5	0.4	2.8 10^{11}	1.15 10^{11}	3 10^{9}	3.5 10^{11}	1.431011	3.75 10^{9}	
Am 243	0.5	0.3	1.23 10^{12}	1.211012	1.12 10^{12}	2. 10^{12}	2 10^{12}	1.86 10^{12}	
Cm 243	0.7	0.5	7.8 10^{11}	6.81 10^{9}	24.3	1.1 10^{12}	9.5 10^{9}	34.3	
Cm 245	0.5	0.3	1.0 10^{10}	1.0 10^{10}	9.4 10^{9}	1.661010	1.66 10^{10}	1.561010	
Cm 246	0.5	0.3	1.66 10^{10}	1.611010	1.431010	2.76 10^{10}	2.67 10^{10}	2.381010	

(*) Large population ALI x 10^{-4} ICRP 30 (1980) and ICRP 61 (1990)

TABLE IX

PERMISSIBLE CONCENTRATIONS AND HAZARD FACTORS OF **LONG LIVED FISSION PRODUCTS IN HLW EXPRESSED PER THM**

Nuclide	Ground & Drinking water limits (*) Bq/l		HAZARD FACTORS l/THM					
			after			after		
	(1980)	(1990)	5 y	200 y	1000 y	5 y	200 y	1000 y
	(ICRP 30)	(ICRP 61)	according ICRP 30			according ICRP 61		
Sr 90	20	6	1.22 10^{14}	1.1810::	6.4 10^{3}	4.08 10^{14}	3.9 10^{12}	2.13 10^{4}
Y 90	20	50	1.22 10^{14}	1.18 10	6.4 10_{8}	4.88 10_{7}	4.72 10^{11}	2.56 10_{7}
Zr 93	500	700	1.32 10^{8}	1.32 10^{8}	1.32 10_{8}	9.4 10^{9}	9.4 10^{7}	9.4 10
Tc 99	1000^{4}	300	4.88 10^{5}	4.88 10^{8}	4.88 10^{5}	1.6 10^{9}	1.6 10_{6}	1.6 10^{9}
Pd 107	10^{4}	3000	3.77 10^{5}	3.77 10^{5}	3.77 10^{5}	1.26 10^{6}	1.26 10_{6}	1.26 10^{6}
Sn 121m	1000	300	5.4 10^{8}	3.6 10^{5}	5.5	1.8 10:	1.2 10_{8}	18.3
Sn 126	100	30	2.0 10^{8}	2. 10^{8}	2. 10^{8}	6.6 10_{8}	6.6 10^{8}	6.6 10^{8}
I 129	2	2	5.77 10_{7}	5.77 10^{8}	5.77 10^{8}	5.77 10_{8}	5.77 10^{8}	5.77 10^{8}
Cs 135	300	100	4.34 10	4.34 10^{7}	4.34 10^{7}	1.3 10^{8}	1.3 10^{8}	1.3 10^{8}
Cs 137	40	10	8.78 10^{13}	9.71 10^{11}	9.15 10^{3}	3.5 10^{14}	3.8 10^{12}	3.66 10^{4}
8a 137								
Sm 151	5.10^{3}	10^{-3}	5.9 10^{9}	5.33 10^{8}	1.12 10^{6}	2.8 '10^{10}	2.6 $\cdot 10^{9}$	5.6 $\cdot 10^{6}$

(*) Large population ALI x 10^{-4} according to ICRP 30 and 61.

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