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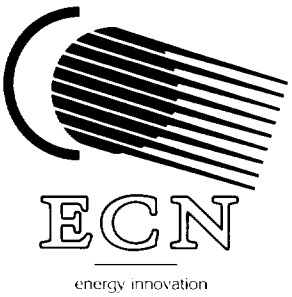

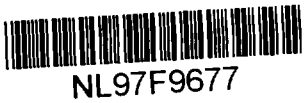

TRANSMUTATION OF NUCLEAR WASTE IN NUCLEAR REACTORS

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SUMMARY

The objective of this joint study of ECN, Belgonucleaire, and Siemens is to investigate possibilities for transmutation of nuclear waste in regular nuclear reactors or in special transmutation devices. Studies of possibilities included the limits and technological development steps which would be needed. Burning plutonium in fast reactors, gas-cooled high- temperature reactors and light water reactors (LWR) have been considered. For minor actinides the transmutation rate mainly depends on the content of the minor actinides in the reactor and to a much less degree on the fact whether one uses a homogeneous system (with the actinides mixed into the fuel) or a heterogeneous system. If one wishes to stabilise the amount of actinides from the present LWRs, about 20 % of all nuclear power would have to be generated in special burner reactors. It turned out that reactor transmutation of fission products would require considerable recycling efforts and that the time needed for a substantial transmutation would be rather long for the presently available levels of the neutron flux.If one would like to design burner systems which can serve more light water reactors, a large effort would be needed and other burners (possibly driven by accelerators) should be considered.

1. INTRODUCTION

For each GWe year generated, the spent fuel from light water reactors contains about 100 kg of long-lived fission products, about 300 kg of radiotoxic actinides (plutonium, neptunium, americium and curium) and 900 kg of residual (either short lived or stable) fission products. During the last fifty years about one thousand metric tons of plutonium has been produced globally. This amount is inherited for about one sixth from the nuclear arms race but most of it originates from commercial nuclear energy production. As this plutonium has an energy content of 800 GWye, it could fuel global electricity for several years at the current level of nuclear energy production.

Plutonium and the minor actinides are about five respectively four orders more radiotoxic than the uranium from which it originated and moreover a large stock of these elements could lead to serious proliferation risks. It seems especially difficult to guarantee that concentrated plutonium will be confined safely for as long as several hundreds of millennia, e.g. because of risks related to human intrusion. It is clear that the plutonium poses the most urgent problem and that reduction of minor actinides is next on the priority list. Because the element neptunium might be rather mobile in most geo-chemical environments, especially the long-lived minor actinide Np-237 and its precursors Am-241 and Pu-241 ought to be taken care of.

In order to reduce leakage dose risks further, the (difficult) large-scale transmutation of the fission products Tc-99 and I-129 is often advocated, especially for the technetium, which is not naturally abundant and in some environments is geo-chemically mobile. Leakage dose risks from these fission products are most probably marginal compared to other dose risks like that from the naturally occurring radium from decay of the U-234 in phosphate fertilizers or from shallow land burial of low level tailings (see table 1 and fig. 1).

High level waste from fuel		
Source	**minimum *)**	**maximum *)**
Transuranium Actinides	10-6 in repositories	in case of massive spilling into ocean or soil between 10-3 and 10-2
fission-products	of the order of 10-6 for repositories	of the order of 10-6 in any scenario
Technologically enhanced background		
Source	**minimum *)**	**maximum *)**
uranium from nuclear fuel cycle	of the order of 10-5 for ocean dump scenarios	3 * 10-4 for massive spilling into the soil
phosphate fertilizers	3 * 10-4	6 * 10-4
*) For each year of current practice, after a supposed cooling-down period of 10 000 years the dose-rate in (arbitrary) units of "present average natural dose-rate"		

Table 1: Comparison of radiation dose-rates on the very long term, whith present dose rates

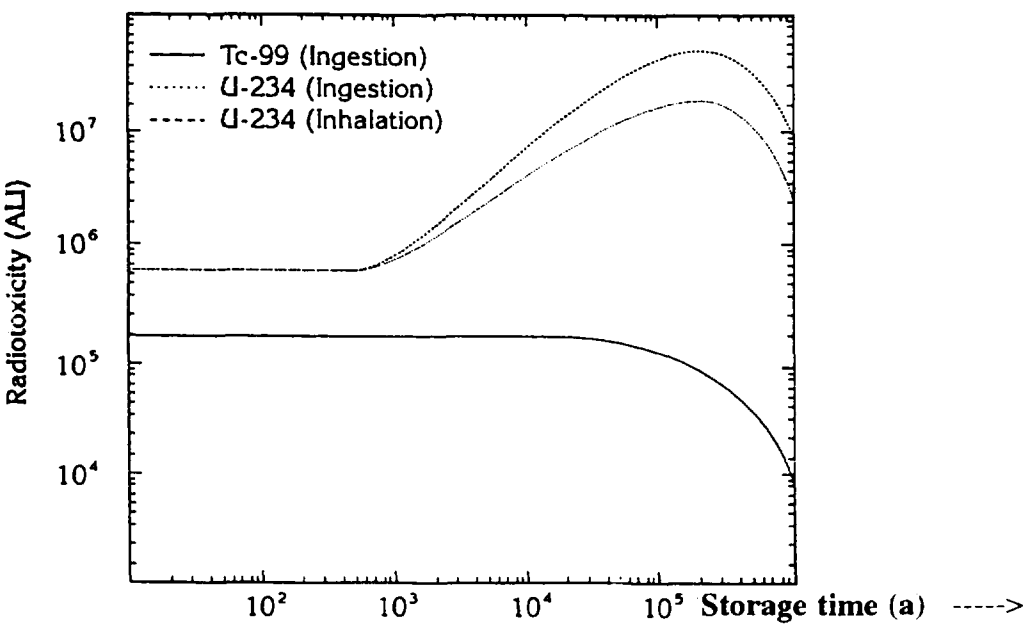

Figure 1: Long term radiotoxicity of Tc and U in spent fuel from a 0.5 GWe year production

On the other hand the radon related risks are still seen as "natural risks" and this can not be said about the technetium risks. If one would like to diminish the marginally small dose risks from remaining technetium and actinide traces, it would seem most worthwhile first to look for compounds which can chemically fix these elements to the storage places for more than hundred thousand years. In case that such an immobilisation can not be guaranteed it should be tried to establish in how far transmutation of these long lived fission products is feasible in a reactor or in a special transmutation device. This report aims at a comparison of methods, by means of the material and conclusions presented in refs 1 and 2. First a common basis for the cross sections used in the transmutation calculations is defined, next the different transmutation devices such as PWRs, FBRs and HTRs are compared and finally a few comments are made regarding accelerator based subcritical reactors for transmutational purposes.

2. CROSS SECTION LIBRARIES and CODE PACKAGES

On behalf of this investigation improved cross section libraries have been prepared and tested by means of various sample calculations. Data libraries of the general purpose fuel depletion code ORIGEN-S (3) have been updated with data based on JEF2.2 and EAF-3. This code uses three-group cross sections in three separate libraries: one for light elements, one for actinides and one for fission products, or alternatively it might use the three libraries combined into one large library with the code COUPLE (4). Another possibility is to use the ORIGEN-S code within the SCALE package and to calculate properly resonance-shielded cross sections within this package. In this way it is possible to average these cross sections over the fuel regions of the reactor of interest; by means of the code COUPLE the one group cross sections are then for any relevant neutron spectrum combined in one binary library.

In total cross sections of 517 light nuclides, 65 actinides and 319 fission products have been renewed or added to the three separate libraries (with cross sections of 604 nuclides). Libraries have been validated by comparing results of calculations for several reactor types; results from regular cross-section updates during the burnup sequence have been compared to results without these updates. For the LWR, the French PWR-N4 reactor was used as a reference design. Cross sections of this reactor type vary much with the burnup.For the LMFBR, the Superphenix reactor was used as a reference design. Also for this reactor the cross sections vary with the burnup, but these variations turned out to be very small.

Physical data, operating history, specific power and initial nuclide densities were given for three different fuel samples with different burnup values of respectively 27.4, 37.1 and 44.3 GWd/tU. Benchmark calculations have been done on three fuel samples for which measurements were available (5). First the proper values for the three spectral parameters were calculated in the discrete-ordinates XSDRNPM code. Secondly, the burnup calculations with the ORIGEN-S code were performed with the three separate cross section libraries. Results of the benchmark with the highest burnup are given in table 2. In general a satisfactory agreement was found and discrepancies between measurements and calculations are within 10 %. It was also found that regular cross section updating leads to improvements which are likewise in the 10 % range. It can be seen that calculations with the new libraries give slightly better results than calculations with the old libraries. Remaining discrepancies for the fission products Se-79 and Sn-126 have still to be cleared up in further work.

Calculations to be treated in the next chapter have also been performed with other codes like the LWR-WIMS code with the WIMS-86 cross section library, with improvements for the Am and Cm isotopes. These calculations have been followed by a second series of calculations based on the JEF2.2 file so that all the work presented in this paper is in accordance with the latest cross section updates and all conclusions will depend on the same data base.

Table 2: *Nuclide concentrations at a burnup of 44.34 GWd/tU in units of mg/g UO_2 and of mCi/g UO_2 for the Burnup Credit Criticality Benchmark part I-B [5].*

Nuclide	Measurement	New Library		Old Library	
		Value	C/E	Value	C/E
U-234	1.20e-01	1.24e-01	1.03	9.03e-02	0.78
U-235	3.54e+00	3.04e+00	0.86	3.20e+00	0.90
U-236	3.69e+00	3.65e+00	0.99	4.03e+00	1.09
U-238	8.25e+02	8.25e+02	1.00	8.24e+02	1.00
Pu-238	2.69e-01	2.27e-01	0.84	2.81e-01	1.05
Pu-239	4.36e+00	3.71e+00	0.85	4.32e+00	0.99
Pu-240	2.54e+00	2.28e+00	0.90	2.18e+00	0.86
Pu-241	1.02e+00	1.03e+00	1.01	8.56e-01	0.84
Pu-242	8.40e-01	8.90e-01	1.06	5.96e-01	0.71
Nuclide	Measurement	Value	C/E	Value	C/E
Np-237*)	3.31e-04	2.91e-04	0.88	4.05e-04	1.22
Am-241*)	1.31e+00	1.17e+00	0.89	9.38e-01	0.72
Se-79*)	6.49e-05	4.30e-04	6.63	4.41e-04	6.80
Sr-90*)	6.58e+01	7.01e+01	1.07	6.87e+01	1.04
Tc-99*)	1.35e-02	1.56e-02	1.16	1.62e-02	1.20
Sn-126*)	2.20e-04	7.22e-04	3.28	7.40e-04	3.36
Cs-135*)	4.95e-04	4.83e-04	0.98	4.43e-04	0.89
Cs-137*)	1.09e+02	1.10e+02	1.01	1.09e+02	1.00

*) These data are given in units of mCi/g UO_2.

On behalf of actual design studies on transmutation of actinides and fission products, one will also need data on the transmutation products, on cladding, and on several matrix materials. Therefore a separate study has been made on the long-range activation cross sections for these materials (6) and this includes a graphical representation of part of the ECNAF data library of cross sections for reactions that lead to the most important reaction products. A total number of 132 graphs of relevant cross sections is given in ref. (6). Not too many surprises have been found in the presented study. An exception could be the long lived Cl-36 activity due to capture in chlorine traces. Even capture of neutrons in Cl-35 from the decay of S-35, the activation product of S-34 might produce some Cl-36 activity.

3. PWR CORE PHYSICS

Recycling of Pu

First of all the reactor assumed for recycling was taken to be a 900-MWe PWR, characterized by a quarter-core fuel management with an average discharge burnup equal to 45 GWd/t. As MOX fuel a design was taken which is equivalent in energy production to UO_2 fuel enriched to 3.7 % in U-235. At discharge all assemblies were considered to have been irradiated for four calendar years at an average load factor of 75 %.

Lead times in the fuel cycle are four years for core irradiation, three years for cooling up to reprocessing, and two years for refabrication and transport before reloading. First a multiple recycling strategy in a reference Pu core (without recycled americium) has been taken as follows:

- In the first stage called MOX 1, the mass of Pu which can be recovered from 1 tonne of UO_2 spent fuel from the initial PWR operation is mixed with UO2 fuel; this corresponds to about one MOX assembly for seven initial UO2 assemblies.
- For the following reprocessing operation it is assumed that all plutonium from one MOX assembly is mixed with the plutonium from six UO2 fuel assemblies. Therefore the plutonium composition available for recycling is a blend, which has a composition different from the composition of the plutonium from the first MOX generation.
- As indicated by the calculations for the next stage MOX there is now isotopically degraded plutonium, which should be blended with the plutonium of about five spent UO2 assemblies, and so on.

The strategy given above is a simulation of what could occur in reality in a whole reactor park, if one would neglect the influence of lag times. A detailed representation of lag times, reactor availabilities and fuel cycle plant pecularities has been made in complementary studies by CEA (7).

As is already indicated in chapter 2 calculations have been performed with the WIMS code. First the data library WIMS-86 has been used, with some improved values for the cross-sections for the isotopes of americium and curium. A second series of calculations used cross-sections from the JEF2.2 file. Salient results for this case of plutonium recyling are presented in table 3, in which one respectively finds the needed Pu enrichments, the

Successive MOX recycling steps	(UO2)	MOX 1	MOX 2	MOX 3
(Pu_{tot} + Am) % in MOX fuel	_	7.8	10.4	11.8
kg of Pu recovered for 1 tonne of heavy metal at latest fabrication	11.0	17.4	22.2	25.9
Isotopic composition of Pu +Am for the next fabrication (%)				
Pu - 238	2.3	3.0	3.6	
Pu - 239	53.2	47.0	44.0	
Pu - 240	24.7	28.2	30.2	
Pu - 241	11.8	12.4	12.4	
Pu - 242	6.8	7.9	8.5	
+ Am-241 from Pu-241 decay	1.3	1.4	1.4	

Table 3: main physics results from multiple recycling of Pu in an LWR

quantities of Pu reprocessed and blended, and the isotopic composition of the Pu available for the refabrication of the MOX fuel. From table 3 it can be seen that the required Pu enrichments grow significantly with the successive recycling steps. This might lead to increasing difficulties in keeping the MOX loadings compatible with the usual design and safety objectives, so that in practice a third step MOX 3, probably has to be the last step in this multiple recycling scheme.

Recycling of Pu and Am simultaneously in LWRs

In pure LWR scenarios the plutonium apparently cannot be recycled more than three times. Such a recycling would only increase the production of minor actinides by neutron capture, unless an extremely high and not yet feasible flux level would have been applied. Nevertheless it is of interest to know the consequences of recycling minor actinides a few times in thermal reactors by admixing these into mixed oxide (MOX) fuel. In this case it is assumed that not only Pu, but also Am can be recovered at spent fuel reprocessing; recycling in the form of MOX fuel (Pu + Am) is considered to take place in the same PWR under the same conditions. A recovery yield of 99.5 % for Pu and 98 % for Am has been assumed.

For the same burnup as in the pure Pu recycling case, the calculational schemes of the above subchapter have been repeated for the sum of Pu and Am, and results are indicated in table 4. This table shows that the enrichments required in this Pu + Am case are still higher than the enrichments needed to recycle only the plutonium. Recycling of the americium by adding it to the plutonium in the MOX fuel would however in practice limit the amount of fuel recycling steps to one instead of three due to the deterioration of the reactivity coefficients. It therefore seems doubtful that a second step MOX 2 will still be acceptable in the Pu + Am case: an additional study, centred on the safety aspects, would be needed to resolve this question of limitation.

Successive MOX recycling steps	(UO2)	MOX 1	MOX 2	MOX 3
(Pu + Am) % in MOX fuel				
Pu	_	9.8	13.6	16.6
Am	_	0.5	1.1	1.6
kg of (Pu + Am) recovered for 1 t of heavy metal at latest fabrication				
Pu	10.9	17.9	23.9	29.2
Am	0.6	1.4	2.3	3.0
Isotopic composition of Pu + Am for the next refabrication (%)				
Pu - 238	2.3	3.5	4.7	
Pu - 239	53.8	48.2	45.7	
Pu - 240	25.1	28.4	30.2	
Pu - 241	11.9	12.2	11.5	
Pu - 242	6.8	7.7	8.0	
Am -241	70.6	64.6	63.7	
Am -242^m	0.1	0.6	1.0	
Am -243	29.3	34.8	35.3	

Table 4: main physics results from multiple recycling of Pu + Am in an LWR

4. FUEL FABRICATION ASPECTS OF Pu + Am RECYCLING IN PWRs

A study has been made on radiation dose protection and criticality safety in a MOX fuel refabrication plant. A major difference with previous studies is that the present work only considers admixtures of americium into plutonium. Other minor actinides were left out in a realistic short term approach; it only had to be assumed that chemical recovery of americium will soon prove to be feasible on a large scale.

As a reference case the MOX fabrication plant of BELGONUCLEAIRE at Dessel (8) has been taken. It was already known that the front end stages are the most critical ones with respect to handling operations on the pure oxide powders. Currently PuO_2 powders from the reprocessors are taken from storage. Subsequently these powders are introduced in a glove box to be milled and blended together with UO2 powders to produce the so-called primary blend. Such dose intensive operations are followed by secondary blending, pressing and sintering, before the sintered pellets are put into fuel pin claddings: dilution first and canning afterwards lower the dose rates. Therefore primary blending is retained as the most typical source of the dose rate due to handling operations.

Neutron and gamma dose rates at a distance of 30 cm from the external glove box wall have been calculated for a given configuration of blending devices (the UO2 silo is not represented), with a glove box and external shielding with a thickness of:

a) 2 mm steel of the silo bottle;

b) 100 mm of polythene sandwiched between two layers of 0.5 mm steel acting as a neutron shield;

c) 10 mm of plexiglass from the glove box wall;

d) 36 mm of " KIOWAGLASS" a composite material, including 30 % of lead.

Note that b) and d) are additional shields which are not present in the standard fabrication line. For the primary blending operation, at maximum 60 kg of oxide can be handled, of which about 30 % can be PuO2 (containing a small amount of Am 241). All calculations have therefore been run with this maximum quantity of 18 kg of PuO2 kept as a constant, but with a variable composition as given in table 3 (Pu case) or table 4 (Pu + Am case). In the latter case AmO_2 is added to these 18 kg of PuO_2. For example the total AmO_2 content is 0.97 kg for MOX 1 (Pu, Am), whereas it was 0.22 kg in the MOX 1 (Pu) case.

Gamma and neutron fluxes are converted into equivalent dose rates by means of standard conversion factors. Standard computing processes have been validated via OECD benchmarks for spent fuel transport and also by comparing results with measurements on MOX sources.

Table 5 gives the dose rates at 30 cm from the glove box calculated for as well the reference PuO_2 powder (MOX 1) as for the various powders, which correspond either to a second Pu recycling step (MOX 2) or to a first recycling of Pu and Am.

A value of 20 micro Sievert per hour (~ 2 mrem/h) is taken as a guiding value for these comparisons, although it is no real limit in the plant as the staff will not stay longer than needed near the glove box of primary blending, according to the ALARA principle.

Table 5 shows that conditions for the MOX 2 fuel fabrication are very similar to those of the MOX 1 fuel. There is indeed only a marginal increase in the main neutron source, which is induced by the alpha radiation from Pu-238 by (alpha, n) reactions. This favourable result is caused by the dilution of the Pu from the MOX fuel with the Pu from UO_2 fuel.

Case	Dose rates (micro Sv / h)		
	Gamma	Neutrons	Total
MOX 1	9.7	7.3	17
MOX 2	9.7	9.1	18.8
Pu Am	67.4	8.0	75.4
Pu Am+20 mm additional steel	17.5	5.8	23. 3

Table 5: impact on MOX fuel fabrication at 30 cm from glove box of primary blending

On the other hand table 5 also shows that the addition of americium at reprocessing induces a strong increase of the gamma dose (with about a factor 4.5 if one assumes a two year delay between reprocessing and fuel fabrication). This increase could be remediated by an addition of shielding with a layer of 25 mm of steel. Table 5 also shows the calculational results if only 20 mm of steel would have been applied. It thus appears that dose rates could be controlled at the expense of relatively little extra shielding efforts. Of course the extra shielding would hinder the fabrication and would increase the cost of operations. Still the operations seem a feasible extension of the standard MOX fabrication conditions especially if one would consider remote fabrication with automated processes.

5. RECYCLING Pu AND Am IN FAST REACTORS

Why choose fast reactors?

As was shown in chapter 3 on PWR core physics, the effect of recovering americium in addition to plutonium at reprocessing and of introducing it in the MOX fuel, is that at the first recycling step already 10 % (Pu + Am) enrichment is needed. In the usual Pu recycling case this is 7.8 %; the 10 % enrichment level is needed at the second recycling step. Recycling Am in addition to Pu in the MOX fuel limits the number of possible recycling steps to one instead of two or three. Actually more refined core calculations would be needed to confirm this limit for a safe core operation.

From the above considerations on fuel refabrication it can be concluded that penalties are similar, whether Pu and Am are recycled once or twice in a PWR, like it has been represented here or whether a large number of cycles are made in fast reactors, like considered in ref (9). It therefore has been suggested that in a longer term fast reactor systems could be more suitable to transmute the actinides, at least if one could limit the deterioration of the sodium void effect and of the Doppler constant.

Plutonium burning in fast reactors

In order to achieve a maximal burning rate, two design measures have to be taken: firstly, the radial and axial breeder zones have to be removed and to be replaced by steel reflectors and, secondly, the internal breeding ratio has to be reduced. This second requirement mainly can be met by increasing the plutonium content, i.e. reducing the U-238 content as new Pu-239 will be formed mainly by capture in U-238. Results of a systematic study are shown in fig.2.

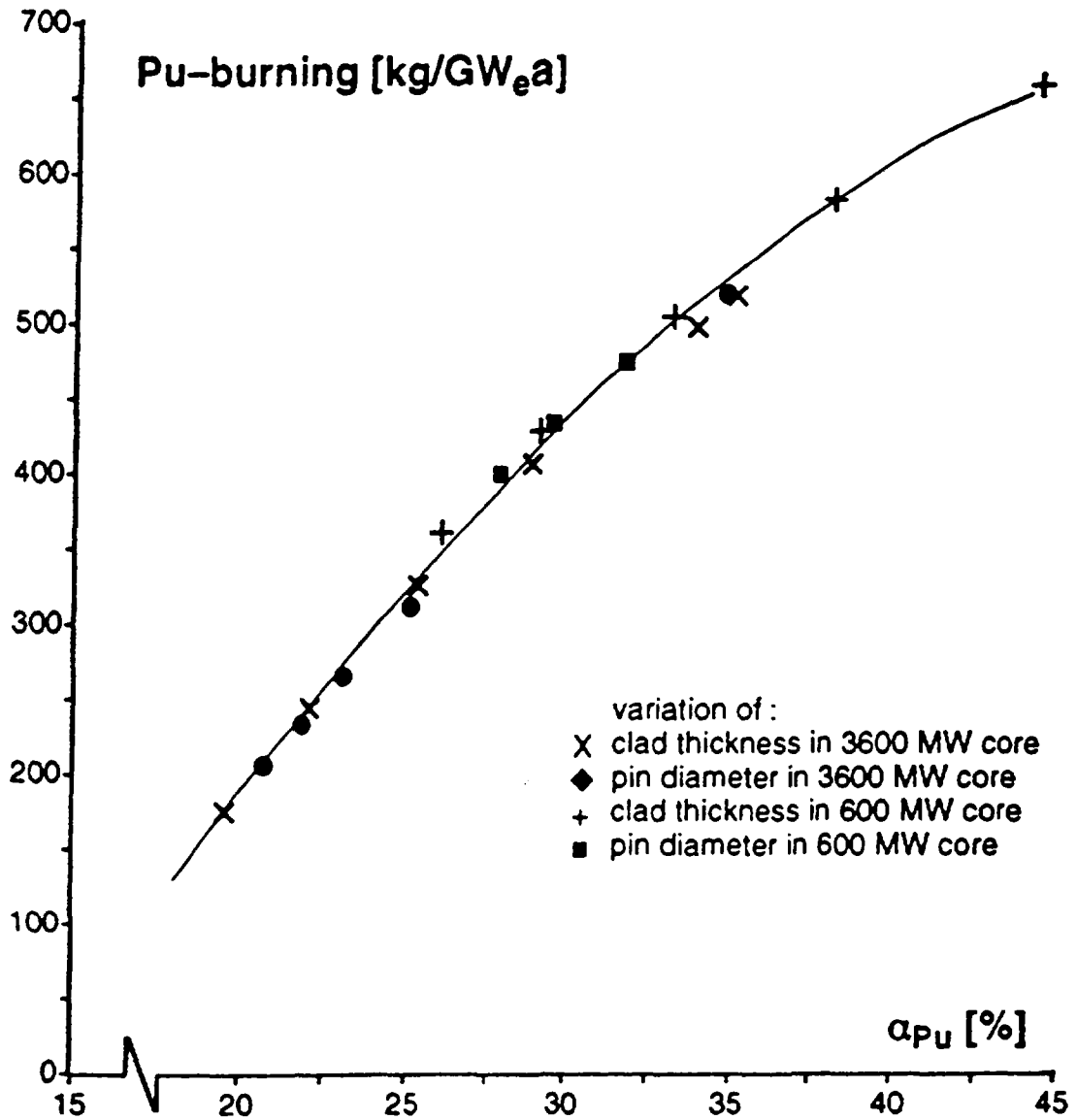

Figure 2: Influence of Plutonium content α_{Pu} on Plutonium burning rate

For a large core with 3600 MWth and a small core with 600 MWth the Pu content was increased by either changing the diameter of the core pins or the clad thickness. Figure 2 shows that the Pu content is the main determining parameter since points of the very different cores fit very well to a single curve.

Up to now experience with respect to behaviour during operation and solubility during reprocessing is available for Pu-contents of up to 30-35 %. For this concentration a Pu burning rate of about 480 kg / GWe a or about 55 kg / TWh can be deduced from fig.2. After further improvements especially with respect to solubility, the limit is expected to be shifted to about 45 %, which would allow a burning rate of almost 700 kg / GWe a or 80 kg / TWh. Such a burning rate would correspond to a Pu production rate of about 4 GWe LWR power.

Conditions for burning of minor actinides

In fast reactors the lanthanides have similar influences to these core properties as the minor actinides. Therefore it would be recommendable to put an upper limit to the total amount of minor actinides and lanthanides together of about 5%. Because the mass ratio of lanthanides and minor actinides in typical spent fuel might be higher than 50, an efficient transmutation of minor actinides would require a chemical separation from the lantanides with a separation factor around 30-50.

In that case also special attention must be paid to the build up of Pu-238 in the fuel, which reaches already a value of 7 % of the total plutonium in case of an admixture of 5 % of minor actinides. Such high amounts of Pu-238 would require very efficient neutron shielding and cooling measures during refabrication of fuel. The short lived Pu-238 would also be a serious waste contaminant. Criteria on the reduction of dose-risks disfavour scenarios in which waste will be contaminated with U-234 or with its precursors. This will be so for the U(Pu) cycle but even more so for the Th (U) cycle, wherein orders of magnitude higher amounts of U-234 will be produced. Geological storage would be needed that prevents the escape of emanations from the waste before the radon has decayed (a few days after it has been formed).

6. PLUTONIUM BURNING IN HIGH TEMPERATURE REACTORS

Basical for this study was the German modular high temperature reactor HTR-M (10). The core of this helium cooled reactor consists of a randomly packed bed of spherical fuel elements of 60 mm diameter, which contain the fuel in the form of coated particles.

The main core parameters of the HTR-M have been kept for this study as:

- Thermal power 200 MW, and average power density 3 MW / m^3

The fuel type has been changed from (U, Pu) to a pure Pu fuel and the Pu composition was taken as a typical weapons-grade one as taken from an American study (11):

- Pu-239 : Pu-240 : Pu-241 = 94 : 5.4 : 0.6

A very high discharge burnup of 700 GWd / t (HM) was assumed, which according to (11) had been succesfully realized in the Peach Botton and Dragon projects. The use of PuO_2 fuel has strong consequences for the safety parameters, especially for the temparature coefficient, which would become positive due to the reduced Doppler coefficient. Therefore erbium-167 is added to the fuel, which besides reducing the temperature coefficient also acts as burnable poison. Cores have been studied with increasing amounts of erbium for which the necessary Pu-content was fixed to obtain the required reactivity.

Fig. 3 shows the evolution of the concentrations of the involved isotopes for a typical case (Er content 14 % of the Pu weight). The Pu-239 continuously decreases, Pu-240 and Pu-241 reach a maximum after four and ten cycles respectively and americium increases continuously.

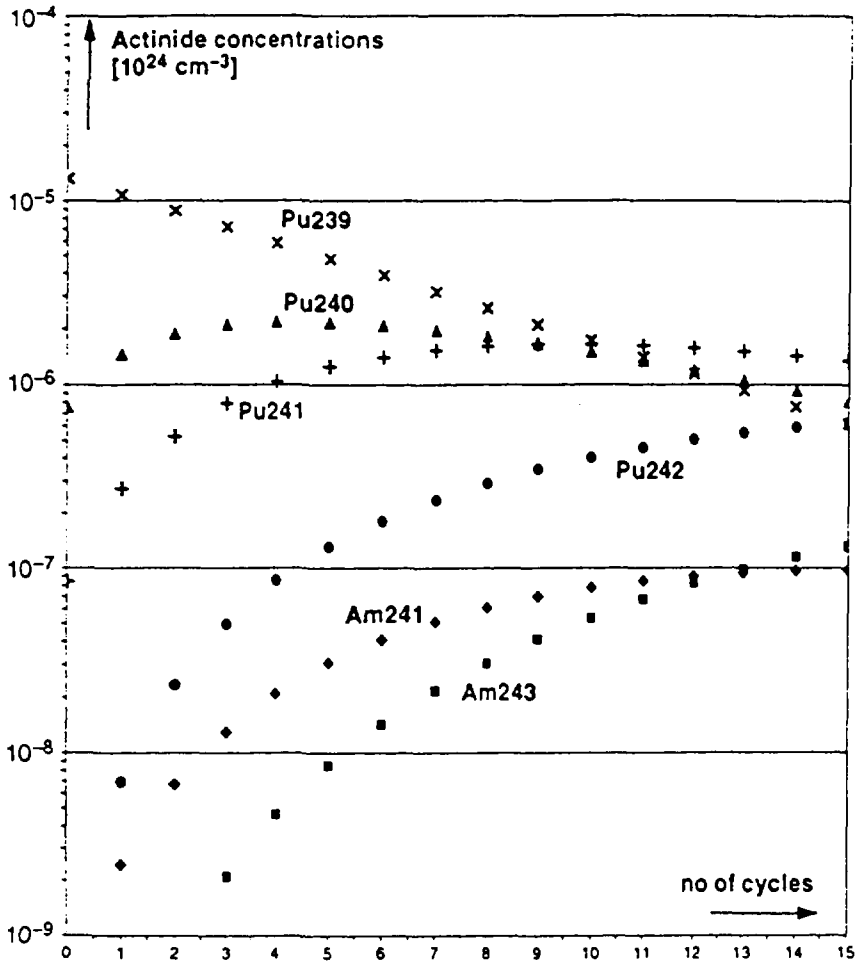

Figure 3: Evolution of the Actinide concentrations during burnup in a HTR with weapons-grade Plutonium

7. RECYCLING LONG-LIVED FISSION PRODUCTS

It has been shown that in the long run the long-lived fission products Tc-99 and I-129 dominate the leakage dose of unperturbed waste repositories. Therefore a study has been made of possibilities to transmute these two geo-chemically mobile elements in current reactors. Heavy water reactors, fast reactors and pressurized water reactors have been considered. It is noted however that not all Tc and I is stored in the form of high level waste, as parts are retained in the reprocessing plant. Only if most Tc and I would be partitioned in concentrated form, with a high decontamination factor, it would make sense to consider transmutation.

For the HWR, a CANDU type with power of 935 MWe as present in Darlington (Can) has been used as a reference. Three cases have been discussed all with an equal initial amount of Tc-99 per fuel bundle and a loaded amount of Tc-99 equal to about 3.8 tonnes, which is quite high. All calculations were performed with the Monte Carlo code KENO (12, 13). It appeared that transmutational half lifes range from 44 years to 25 years. It turned out to be best to position the technetium pins in the moderator between the fuel bundles. With a gross transmutation rate of 100 kg/a, and a new HWR production of 20 kg/a, the nett transmutation rate would be about 80 kg/a, and then one HWR could serve four LWRs with equal power.

Irradiation of Tc-99 and I-129 in standard light-water reactors has been considered as well. Special pins are supposed to be placed on the guide tube positions of a 900 MWe PWR core. It has been assumed that these pins would contain either Tc-99 metal or I-129 diluted with the natural I-127 in a ratio one to four and in the form of cerium iodide (CeI_3).

Neutron physics calculations aimed first at determining an acceptable loading so that the operation of the reactor is not penalized, next the efficiency of transmutation has been derived. At first the calculations were performed with the WIMS-86 cross-section package. A second series used the more recent JEF-2 file. Although the results changed slightly, these calculations have broadly confirmed each other.

Two cases have been considered for the driver fuel, which was either UO_2 (enriched 3.7 % in U-235) or MOXwith an equivalent Pu content so as to reach a burnup of 45 GWd/kg. Although the transmutation is better in UO_2, a MOX loading can more easily be adjusted.

In current LWRs one would like to irradiate at a low density (about 1 kg technetium per liter in an inert matrix). In this case a transmutation rate of about 25 kg per year in a total inventory of about 750 kg technetium in guide tubes in the reactor would be reached. A break even situation (Tc-99 transmutation rate equal to its production rate) cannot easily be achieved because in practice not enough guide tubes will be available for transmutation purposes. Transmutation efficiencies are always modest. In the UO_2 case one reactor loading can transmute the Tc production of 1.6 reactors of the same type, so that two PWRs on three should be loaded with the envisaged Tc targets to ensure about equilibrium. For I-129 the necessary ratio should be two PWRs on five.

For calculations on transmutation of Tc-99 in a fast reactor, the Superphenix reactor with a thermal power of 3000 MW was used as a reference design. It was assumed that the Tc-99 target pins are irradiated in a special moderated subassembly and in a special non-moderated subassembly, both loaded in the inner core of Superphenix. In order to increase the transmutation rate the amount of target material should be optimized. In this case some extra fuel enrichmrent is needed. In this case the gross Tc-99 transmutation rate might reach 100 kg/a and four 1000 MWe LWRs could be served. The transmutational half lives are between 15 and 20 years.

8. TRANSMUTATION BY MEANS OF ACCELERATOR DRIVEN SYSTEMS

In order to reduce the transmutational half lifes and to be able to work with smaller inventories, one would like to use special high flux systems driven by accelerators. If a beam of GeV protons hits a spallation target, many neutrons will be released via spallation and evaporation processes. The number of fissions afterwards then depends additionally on the effective multiplication factor k_{eff} of the whole system. Literature on the study of different spallation aided systems and their intercomparison has shown that these systems can be very efficient as a transmutation device for fission products as well as for actinides. If one uses an external neutron source, the reactor can be in a sub-critical state in which some requirements for safety may be relaxed. Consequently one may work with a system at a high neutron flux, which has temperature- and void- reactivity coefficients, which would not be allowed otherwise. Thus the concentration of actinide waste might be increased beyond the limits imposed earlier (5 % for minor actinides), and one transmuter may serve more LWRs than the earlier derived optimum of about five if this concentration can be increased. But also for the fission products the concentration in the reactor could be increased strongly in a subcritical system. For fission products this is not so much thanks to a relaxation of safety requirements as well as because of the requirement of reaching criticallity under condition of extra neutron absorption.

For sodium cooled fast reactor systems (like PHOENIX and a JAERI concept) the transmutation rate per GWe produced can be increased by more than a factor ten for minor actinides. A thermal system as the Los Alamos concept ATW will not do as well, but this system is more advocated to transmute technetium and military plutonium. This Accelerator Transmutation of Waste concept is either a low k_{eff} design, which is a product of previous military investments in an Accelerator Tritium Production system (ATP), or a high flux ATW with k_{eff} up to 0.95.

In general there are three points of concern if one compares sub-critical accelerator-driven systems with critical reactors for transmutation. First of all the economy of an accelerator driven system will be reduced, because a fraction of the electric power generated in the reactor will have to be sent back to the accelerator instead of to the grid. This fraction is proportional to $(1/k_{eff} - 1)$, and if k_{eff} is too small, waste transmutation might easily cost more electricity than has been produced in the course of generating the waste. Such a situation could be possible for military waste, but is not likely for commercially produced waste from the electronuclear energy production. Secondly the transmutation system might produce its own waste, which should be taken care of. As soon as the minor actinide content increases beyond a few percent a large fraction of Pu-238 will be formed. Problems due to Pu -238 have been treated in the sub-chapter on minor actinide transmutation. Last but not least one should work on the cross sections at high energy: although the uncertainties due to inconsistent libraries of cross sections were shown to be relatively small at low energies, this is certainly not the case at high energies.

Several other aspects would still have to be clarified, in particular regarding the feasibility of the intense high-energy accelerator beam in a non-pulsed operation regime and regarding the claimed safety advantages of high power sub-critical systems. Refabrication of fuel and the material problems linked with the high-energy radiation would have to be taken care of as well.

9. CONCLUSIONS

In this joint study ECN, Belgonucleaire, and Siemens investigated possibilities for transmutation of nuclear waste in regular nuclear reactors or in systems based on advanced

special transmutation devices. On behalf of this investigation improved cross section libraries have been prepared. Next the burning of plutonium in fast reactors, gas-cooled high-temperature reactors and light water reactors (LWR) has been considered. Finally the transmutation of minor actinides and fission products has been studied.

It has been shown that for pure LWR scenarios the plutonium cannot be recycled more than three times. Such a recycling would only increase the production of minor actinides by neutron capture, unless an extremely high and not yet feasible flux level would have been applied. A study has been made on radiation dose protection and criticality safety in a MOX fuel refabrication plant. A major difference with previous studies is that the present work only considers admixtures of americium into plutonium. It appeared that dose rates could be controlled at the expense of shielding efforts. Recycling of the americium by adding it to the plutonium in the MOX fuel would however in practice limit the amount of fuel recycling steps to one instead of three due to the detoriation of the reactivity coefficients.

In a longer term fast reactor systems will be more suitable than LWRs to transmute the actinides, at least if the deterioration of sodium void effect and Doppler constant are limited. In fast reactors lanthanides have similar influences to such properties as minor actinides, and it would be recommendable to put an upper limit to the total amount of minor actinides and lanthanides together of about 5%. Because the mass ratio of lanthanides and actinides in typical spent fuel might be higher than 50, an efficient transmutation of minor actinides requires a chemical separation from the lantanides with a separation factor around 30 to 50.

Another result of the study was that for a well designed and safe system the transmutation rate for minor actinides mainly depends on the content of the minor actinides in the reactor and to a much less degree on the fact whether one used a homogeneous system (with the actinides mixed into the fuel) or a heterogeneous system (with the actinides in seperate sub-assemblies embedded in an inert matrix material). A natural limit for the actinide destruction capability of critical reactor systems seems to exist. So if one wishes to stabilise the amount of actinides from LWRs, about 20 % of all nuclear power would have to be generated in special burner reactors. If one would like to design burner systems which can serve more light water reactors, a large effort would be needed and special purpose burners (possibly driven by accelerators) should be considered.

In case of minor actinide transmutation, special attention must be paid to the build up of Pu-238 in the fuel, which reaches already a value of 7 % of the total plutonium in case of an admixture of 5 % of minor actinides. Such high amounts of Pu-238 would require very efficient shielding and cooling measures during reprocessing and during refabrication of fuel. The short lived Pu-238 would be a serious waste contaminant. Criteria on the reduction of dose-risks disfavour scenarios in which waste will be contaminated with U-234 or with its precursors. This will be so for the U(Pu) cycle but even more so for the Th (U) cycle, wherein orders of magnitude higher amounts of U-234 will be produced. Geological storage that prevents the escape of emanations from the waste before the radon has decayed (a few days after it has been formed) would be needed for such waste.

In order to reduce leakage dose risks further, the (difficult) large-scale transmutation of the fission products Tc-99 and I-129 could be taken up, especially for the technetium, which is not naturally abundant. Dose risks from this fission product are however marginal, and efforts related to a complete recycling of all technetium, would be considerable. It can therefore be concluded that in this way the technetium problem is probably not to be solved at a price worth the benefits. Efforts to reduce the leakage of technitium by chemically fixing this element in a geologically stable storage position, therefore deserve a higher place on the priority list than efforts directed to partioning and transmutation.

Literature on the study of different spallation systems and their intercomparison has shown that these systems can be very efficient as a transmutation device for fission products as well as for actinides. However several aspects would still have to be clarified.

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