IMPACT OF HIGH BURNUP IRRADIATION AND MULTIPLE RECYCLE OF PLUTONIUM AND MINOR ACTINIDES ON THE FUEL CYCLE ACTIVITIES

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ABSTRACT

The paper shows the impact of increasing burnups and recycle of MOX fuel in thermal and fast reactors. Reprocessing of the targets for multiple recyling will become increasingly difficult as the burnup increases from 4 to 7 At % in LWR-MOX fuel and 12-20 % in fast reactor fuel.

Multiple recycling of Pu + MA in fast reactors is a feasable option which has to be studied very carefully: Pu, Am and Np levels decrease as a function of the recycle number, Cm accumulates and gradually transforms into Cm-245, 246 and 247. Long cooling times are necessary with aqueous processing.

At each recycle step PUREX reprocessing will produce a concentrated HLW solution which contains all the fission products, minor actinides and U-Pu residues. These solutions are the source terms for partitioning operations.

The presently developed partition operations DIDPA, TRUEX, DIAMEX and TRPO do extract quantitatively the actinides from the concentrated waste solution. The Am-Cm fraction is a mixed actinide-lanthanide product stream which has to be treated for partial lanthanide removal.

New fuel fabrication technologies will have to be developed in order to cope with increased Pu-238 levels. The preparation of fuel or targets containing Am-Cm is the biggest challenge to be overcome.

1. Introduction

The recent evolution in the nuclear fuel cycle management is influenced by a number of important technical and political facts which have changed the traditional outlook of this nuclear activity.

- There has been a general tendency in reactor fuel management to increase the burnup of irradiated UO_2 fuel from 33 to 43 Gwd/T and even burnups beyond that value are considered.

- The reprocessing capacity has doubled but is not yet capable of keeping pace with the production of spent fuel, increasing furthermore the backlog of spent fuel. A number of countries have modified their position towards reprocessing as reference fuel cycle option.

- The use of separated Plutonium has shifted from Fast Breeder Reactors to Light Water Reactors. The potential market for LWR-MOX fuel is very large but depends on the availability of LWR-MOX fuel fabrication plant-capacity.

- The quantity of separated Plutonium exceeds the present usage trend and the potental arrival of high quality Plutonium from military origin with an outspoken proliferation risk puts the emphasis on Plutonium rather than on Minor Actinides.

- Due to technical and institutional reasons the geologic disposal approach has further been shifted into the future and there is time to develop technologies to decrease the potential toxicity of Actinides and certain fission products.

2. Evolution of quantities of actinides and their radiotoxicity as a function of burnup

2.1. UO_2 fuel

Most of the earlier radiotoxicity - and actinide - inventory calculations were made on the basis of UO_2 fuel irradiated up to 33 GWd/T. [1], [2].

This burnup rate has since long time been surpassed by operational power plants. The present standard burnups are close to 43 GWd/T and this results in an increase of the fuel irradiation time, an increase in actinide generation per unit of weight of fuel and an increased residual heat. By doing so the specific fuel cycle costs, the residual Pu and the residual fissile contents decrease sharply. Table 1 gives an overview of this tendency.

Table 1
Influence of increasing burnup on Pu inventory of UO_2 fuel [3]

Burnup GWd/THM	THM/GWe-year (*)	kg Pu/THM	Annual Plutonium inventory		
			Total kg/GWe	Fiss. %	Fissile kg/GWe
33	33.5	9.17	307	69	212
40	27.6	10.24	283	66.5	188
50	22.1	10.86	240	63.2	151
60	18.4	11.74	216	58.8	127
(*) 33 % electric yield - to be multiplied with the load factor (0.75 to 0.85) to obtain net values per year.					

With a load factor of 80 % the total Pu inventories range from 246 to 173 kg Pu/GWe - 0.8 year. The radiotoxicity of the discharged spent fuel does not follow this trend due to an important increase of the Pu-238 and Cm-244 content of this fuel. Table 2 shows the evolution of the radiotoxicity as far as this property is equated to the α content of the fuel.

Table 2
Radiotoxic inventory of highly irradiated UO_2 fuel in Ci α/THM
7 years after discharge from the reactor

Burnup GWd/THM	Plutonium		Np-237	Am-241	Cm-244	α Total
	Total	Pu-238				
33	3 213	2 360	0.15	1 310	1 490	6 050
40	4 430	3 480	0.40	1 500	2 700	8 750
50	6 720	5 740	0.51	1 950	5 700	14 300
60	9 270	8 220	0.64	2 430	11 600	23 500

By comparing Table 1 with Table 2 it is striking that while the total Pu content is decreasing the radiotoxicity increases when expressed in total α per GWe year.

Burnup	Total Ci α / GWe-year	Net Ci α / 0.8 GWe-year
33	$2.02\ 10^5$	$1.61\ 10^5$
40	$2.41\ 10^5$	$1.92\ 10^5$
50	$3.16\ 10^5$	$2.52\ 10^5$
60	$4.31\ 10^5$	$3.45\ 10^5$

There is a slightly more than linear increase in the total α activity released per GWe-year and 80 % of this value per installed GWe for a mean load factor of 80 %.

The reprocessing plants which have to process high burnup fuel will have to cope with a steadily increasing α toxicity due essentially to Cm-244, Pu-238 and to a lesser extent to Am-241. The half-lives of these radionuclides are respectively 18.1, 87.7 and 432 years. Delayed reprocessing will only partially influence the Cm-244 contribution, the other two strong α emitters do not decay appreciably within a technological time frame of e.g. 20-30 years.

2.2. MOX fuel

Recycle of recovered Pu from reprocessed UO_2 fuel has shifted from FBR's to LWR's. The industrial recovery of plutonium from MAGNOX (low burnup) and more recently from UO_2 fuel (33 GWe/T) has led to the creation of a considerable stock of separated plutonium. According to well informed agencies (OECD, IAEA) this stockpile amounted to 92 THM Pu at the end of 1992.

The large reprocessing units (La Hague, THORP) produce together 28 THM Pu/year at full capacity. The degradation of the plutonium spectrum due to the decay of Pu-241 into Am-241 is the main drawback for keeping a stockpile unused apart from the economic penalties.

The typical isotopic composition of different types of plutonium used industrially at the MOX fuel fabrication plant of BELGONUCLEAIRE (Dessel, Belgium) are shown in Table 3.

Table 3
Typical isotopic compositions of plutonium processed at BN for making MOX fuel

Isotope	A	B (gas-graphite reactor)	C (LWR)	D (LWR)	E (LWR recycled)
Pu-238	0	0.05	0.5	1.5	2.3
Pu-239	94.0	79.0	43.1	58.5	48.6
Pu-240	5.45	17.0	40.0	25.0	27.3
Pu-241	0.50	3.5	5.9	10.0	14.8
Pu-242	0.05	0.45	10.5	5.0	7.0

The incentive to use the separated plutonium in LWR-MOX is very high since this is at the present time the main outlet for plutonium. The previous application in fast breeder reactors is presently at a very low gear and the facilities have been either closed or reoriented to LWR or Pu incineration purposes.

By blending plutonium from different origins it is possible to prepare MOX fuel with a reasonable high fissile plutonium content (65-70 %). The potential occurrence of excess plutonium from military origin, with its very high fissile content (Table 3, type A), on the nuclear fuel market would greatly improve the flexibility of MOX use in LWR's and increase the recycle frequency and the time span during which plutonium would contribute to the LWR-fuel cycle.

Two nuclides are limiting the use of recycled plutonium in fresh MOX fuel: Pu-238 and Am-241. In manually operated MOX fuel fabrication plants a concentration level of 1.5 % Pu-238 is acceptable; above that level special neutron shielding has been installed.

The Am-241 content depends on the initial Pu-241 content and the time interval between reprocessing and fuel fabrication. There is no technical procedure to influence the ingrowth once the PuO_2 powder has been delivered to the fuel fabrication plant. The unbalance between the plutonium output of the large LWR-UO_2 reprocessing plants (La Hague, THORP) and the available fuel fabrication facilities is undoubtely the most severe constraint on the industrial use of MOX fuel.

Two strategies could be pursued to solve this serious problem:
- develop Pu purification units to remove Am-241 from old plutonium stocks,
- design and operate fully shielded and remote handled MOX fuel fabrication facilities.

In the first option a solution can be found for the use of old plutonium stocks. The second option is much more difficult to realize but offers perspectives for the production of MOX fuel with a significant minor actinide content. The economic and radiological impact of each of the two options will determine which one has most merits to be implemented on industrial scale.

Because of the increased fuel fabrication costs the irradiation of MOX fuels to higher burnups was asumed from the beginning. Burnups of 40 to 60 GWd/T are currently being considered.

Since the aimed burnup determines the necessary initial Pu $_{Fiss.}$ enrichment and as the Pu $_{Fiss.}$ content of the discharged UO_2 fuel varies acording to its burnup and cooling time (see Table 1), this complex interaction will result in a total plutonium concentration which generally varies from 6 to 8 % Pu total.

After irradiation the composition of spent MOX fuel is approximately shown in Table 4.

Table 4
Indicative spent MOX fuel composition (7 years cooling) [4]

Burnup GWd/T	33	40	50
Initial total Pu %	5.03	5.58	5.58
End concentration Pu %	3.56	3.8	3.50
Plutonium isotopic composition % after 7 years			
Pu-238	2.7	2.6	3.0
Pu-239	39.0	40.8	37.4
Pu-240	34.0	32.4	32.8
Pu-241	14.5	13.6	13.8
Pu-242	9.8	10.3	12.7
Minor Actinide content kg/THM			
Am-241	1.95	2.56	2.42
Am-243	1.07	1.05	1.28
Cm-244	0.37	0.428	0.635
Cm-245	0.03	0.05	0.08

These data are only approximative and need further certification since the initial composition plays a very important role in the production rate of the heavy minor actinides. Experimental verification of the actinide composition based on post irradiative analysis of discharged MOX fuel with a very good irradiation history should be carried out in order to quantify with more accuracy the most critical nuclides: the plutonium isotopes in particular Pu-238, the minor actinides especially Am-241 and Cm-244. The ARIANE programme proposed by BELGONUCLEAIRE is a very good initiative in this direction.

Nevertheless a rough calculation based on these actinide yields shows that the overall α-load from MOX fuel is much higher than that of UO_2 fuel at the same burnup. Table 5 shows the global values.

Table 5
Radiotoxic inventory of highly irradiated MOX fuel in Ci α/THM 7 years after discharge

Burnup	Plutonium		Minor Actinides		Total α load	
GWd/THM	Total	Pu-238	Am-241	Cm-244	Total α	Total/GWe-y
40	2.07 10^4	1.69 10^4	8.70 10^3	3.45 10^4	6.40 10^4	1.76 $10^{\S}$
50	1.92 10^4	1.81 10^4	8.27 10^3	5.12 10^4	7.90 10^4	1.75 10^6

Expressed per 1 GWe year discharged fuel the α radioactivity to be handled in a conventional reprocessing facility is about 5 to 7 times higher with MOX than with the corresponding UO_2 fuel as can be deduced from comparing the data of Table 5 with those of Table 2.

However in the near future only 30 % of the reactor core will be loaded with MOX fuel. By "diluting" 1 MOX fuel element with 2 UO_2 fuel elements the increase of the α radioactivity load is reduced to a factor 2 to 3.5 in the reprocessing plant operations compared to the equivalent LWR UO_2 fuel.

3. Source term evolution in multiple recyling

3.1. Recycle of UO_2-fuel

The possible recycle of reprocessed U (U_{Repro}) has been discussed in an earlier paper [5]. The most important conclusion yet, is that in the present economic circumstances only a small fraction will be recycled. With a majour U_{Repro} stream as waste, instead of product, the separation of Np-237 from HLW cannot be justified on the basis of a radiotoxicity assessment because U-234 and U-236 are present in U_{Repro}.

The HLW solution resulting from LWR-UO_2 fuel reprocessing is the first source term for minor actinide partitioning. Quantitative separation of the Am-Cm fraction before vitrification would result in a substantial decrease of the residual toxicity of vitrified HLW.
By systematically recycling the plutonium, occluded in the spent LWR-UO_2 fuel we achieve a first "concentration" of the actinides' mass with a factor of 5 at least.

After irradiation the total Pu content has furthermore been reduced by 25 to 30 % depending on the burnup but the spent MOX fuel becomes a 3 to 5 times more concentrated actinide source term. If no reprocessing of MOX fuel is envisaged in the near future on an industrial scale it is the first intermediate actinide source term which has to be taken into account in the comparative evaluation of the long term toxicity of different scenario's. If industrial reprocessing of MOX fuel is considered in a medium term the resulting Pu product will become more enriched in Pu-238 and the fuel fabrication technology will have to be adapted due to increased α activity, heat and neutron emission.

3.2. Recycle of MOX fuel in LWR's

Recycle of MOX fuel in LWR's has been investigated for self generated plutonium by WIESE [4]. The fissile Pu content decreases from 63 % in fresh MOX to 48.7 % in spent MOX fuel. This partially depleted plutonium is subsequently mixed with fresh Pu from LWR-UO_2 reprocessing at each recycle. By following this approach multiple recycling of MOX is not excluded if the fissile Pu content is gradually increased to counter effect the steady increase of Pu-242 up to 25 %. The total Pu enrichment must in this hypothesis be increased from 6.32 % in the first recycle to 8.9 % at the 5th recycle with an equivalent fissile Pu content of 3.7 to 4.4 %.

However the minor actinide radioactive inventory resulting from the repeated reprocessing of spent MOX recycle fuel increases very sharply especially Cm-244. From an analysis of the above published data it appears that the Pu consumption increases from 20.8 kg/THM to 26.6 kg/THM at the 5th recycle. However the minor actinides are produced at a rate of 4.8 kg in first generation MOX to 8.9 kg/THM at the 5th recycle. There is a net total actinides consumption of ~ 100 kg in 6 recycles per THM.

By examining the radiotoxicity evolution it appears that the decrease of Pu-238 and the increase of Am-241 are more or less in equilibrium as shown in Table 6, but the generation of Cm-244 is the overwhelming phenomenon.

Table 6
Radiotoxicity evolution of MOX fuel (50 GWd/T) in repeated recycling (Ci/T) after 7 years

	MOX	1st recycle	2nd recycle	3rd recycle	4th recycle	5th recycle
Pu-238 (x 10^3)	+ 0.87	- 5.62	- 8.7	- 9.8	- 11.7	- 12.6
Am-241 (x 10^3)	+ 8.2	+ 9.5	+ 10.7	+ 12	+ 12.7	+ 13.4
Am-243 (x 10^3)	+ 0.25	+ 0.35	+ 0.42	+ 0.51	+ 0.56	+ 0.62
Cm-244 (x 10^3)	+ 51.3	+ 69.4	+ 82.2	+ 95	+ 102	+ 110
Ci α-Balance (x 10^3) per THM	+ 60.7	+ 73.6	+ 85	+ 97	+ 103	+ 112
Heat emission KW/THM due to Cm-244	1.7	2.35	(2.8)	3.23	(3.5)	3.76

3.3. Recycle in fast reactors

As the recycle in LWR-MOX gave rise to a very much increased radiotoxicity and to a disturbing Cm-244 concentration level it was examined what could be obtained by recycling in fast burner reactors. A computation was performed with the ORIGEN-2 programme to assess the effect of increased burnup per cycle and to investigate multiple recycling of LWR-MOX spent fuel in a fast reactor core.

The initial conditions of loaded fuel were as follows:
67 % depleted UO$_2$
33 % Trans Uranium Elements:

29.4 % Pu		3.6 % Minor Actinides (MA)	
% Pu isotopic composition		% MA composition	
Pu-238	3.08	Np-237	5.09
Pu-239	37.25	Am-241	49
Pu-240	33.62	Am-243	30.8
Pu-241	17.1	Cm-244	15.18
Pu-242	8.95		

Two levels of mean average flux were considered:
3.8 10^{15} n cm^{-2} s^{-1} to reach a burnup of 200 MWd/T
2.44 10^{15} n cm^{-2} s^{-1} to reach a burnup of 129 MWd/T after a reactor cycle operation of 1846 EFPD (~ 5 years).

The single cycle with a burnup of 200 MWd/T resulted in a Pu inventory drop of 26.6 %. The values for the minor actinides varied in burnout: Am 27.5 %, Np 30.5 %, whereas the Cm level increased with 63.4 %.

For the more conservative burnup of 129 MWd/T a multiple recycle scenario with 4 cycles was investigated. Each of the cycles lasted for 1846 EFPD and between each cycle a cooling time of 10 years was taken into account. The results of this calculation are summarized in Table 7.

Table 7
% Depletion of TRU's by multiple recycling in a fast reactor

4 cycles of 1846 EFPD ; Intermediate cooling time: 10 years ;
Burnup per cycle: 129 MWd/T; Flux: 2.44 10^{15} n cm^{-2} s^{-1}

	Cycle 1	Cooling 1	Cycle 2	Cooling 2	Cycle 3	Cooling 3	Cycle 4	Cooling 4
Pu	19.5	21.7	14.1	15.3	13.2	14.3	13.2	14.3
Am	11.4	-17	29	16.1	29.8	17.7	29.21	16.5
Np	21	12	22	13.3	23.1	16.3	23	17.15
Cm(*)	-44.3	4.4	-42	3.8	-39	4.1	-34.6	5.7
(*) negative depletion is enrichment.								

By combining the individual depletions per cycle we obtain a global residual inventory of
50 % for Pu
47 % for Am
47 % for Np
83 % for Cm.

Whereas an actinide burnup of 129 MWd/T constitutes an overall depletion of 54.5 % in 4 cycles, there is a good correlation between the overall actinide consumption due to burnup and the individual nuclide depletions except for Cm. It is however striking that the 10 years cooling time between each cycle is particularly effective for Cm depletion ; without cooling periods the inventory would have increased by a factor of 3.75.

The above results have to be analyzed from radiotoxicological point of view and be investigated for each important α emitter i.e. Pu-238, Am-241, Am-243 and Cm-244.

The results of this computation are shown in Table 8 which shows for the most significant nuclides the inventory of α emitters in Ci/THM.

Table 8
Radiotoxicity of recycled MOX fuel in fast reactors after 10 years cooling, expressed in Ci α/THM (same conditions of Table 7)

	Cycle			
Elements	1	2	3	4
Pu-(238, 239 240)	1.8 10^5	1.59 10^5	1.35 10^5	1.07 10^5
Am-(241, 243)	0.76 10^5	0.59 10^5	0.43 10^5	0.3 10^5
Cm-(244)	3.25 10^5	2.82 10^5	2.54 10^5	1.95 10^5
Totals	5.8 10^5	5.0 10^5	4.3 10^5	3.32 10^5
Relative toxicity versus initial charge	0.84	0.72	0.62	0.48

In the short terme toxicity, which is considered here, Np-237, Pu-242 and Cm-245 ... do not contribute in any significant manner. The main conclusion is that the radiotoxicity of the MOX-mixture actinides, is reduced in the same proportion as the global TRU inventory. Cm-244 still plays an important role but does not increase with recycling as it is the case for LWR-MOX recycle (see Table 6). Fast reactor iradiation is an adequate way to reduce the actinide inventory but it takes a long time to achieve a significant reduction. Indeed the reactor irradiation takes 5 years, cooling time 10 years and processing 2 years. Globally each cycle would last 17 years and the actinide reduction obtained is 50 % after 4 cycles (68 years). One way to speed up the incineration pace is by increasing the burnup. The above mentioned fuel inventory would have been reduced to 30 % if a burnup of 200 MWd/t would have been reached in the fuel. However this limit of the attainable burnup depends on the cladding material of the fuel pins and the specific power output of the irradiated fuel. Since multiple recyling involves multiple reprocessing it has to be examined which burnup level is acceptable for aqueous processing.

4. Status of multiple recycle fuel processing

The residual power released by spent fuel is determined by the TRU content, the burnup and the cooling time. Reprocessing of LWR-MOX fuel after 7 years cooling has to cope with an increased residual heat load compared to LWR-UO_2. Standard 33 GWd/T- LWR-UO_2 fuel has an heat output of 1.3 KW/THM, this value increases to 3.27 KW/THM for LWR-MOX fuel of 40 Gwd/T and 4.16 KW/THM at 50 Gwd/T. As mentioned in section 2.2, the impact of this increased heat generation has generally been mitigated by dilution of LWR-MOX fuel with equivalent LWR-UO_2 fuel.

With fast reactor fuel the heat problem is much more important even with a cooling period of 10 years: the increased burnup (to e.g. 180 GWd/T) results in a 7 KW/THM heat generation due to the fission products; the high concnetration levels of Pu-238 and Cm-244 produce an additional heat load of 14 KW/THM. Due to this very high decay heat of ~ 21 KW/.THM spent fuel, full size fuel assemblies cannot be chopped in present generation bundle shears. Up to now spent FR-MOX fuel assemblies are disassembled prior to individual pin chopping or multi pin chopping. Some tons of FR-MOX fuel with a burnup up to 180 Gwd/T have nonetheless been reprocessed successfully at Dounreay and Marcoule. The radiation resistance of the conventional extractant TBP is very well known and well understood. The solvent withstands irradiation doses of 30 Wh/l if the contact times are short (pulse columns, centrigugal extractors). The conventional separation U Pu/Fiss. product + actinides can be performed with aqueous processing methods but the solvent will have to be washed more intensively.

Once the raffinate is separated and the so called HAW is sent subsequently to the acid recovery and the waste evaporator the specific heat problem becomes more stringent.

Indeed the volume reduction between HAW and its concentrate HAWC is generally a factor of 10-15 i.e. the ~ 15 KW/THM decay heat is confined within a 350 to 500 l volume which would correspond to 30 W/l or more. All operations on this type of effluent will be difficult unless sufficient dilution is provided before a chemical process is started. It would be preferable to carry out the partitioning processes on the HAW raffinate before its concentration to HAWC, but his approach can only be realized in a, on purpose, designed waste processing and acid recovery system.

If shorter cooling times are to be considered new separation methods will have to be envisaged. Pyrometallurgical methods might be used as first rough separation technique.

5. Critical overview of the actinide partitioning methods [6]

There are four aqueous methods which have up to now been developed for the separation of actinides from high Active Waste Concentrate (HAWC).

The best known method is the american TRUEX process based on the extractant CMPO, the second one is the japanese DIDPA method, the third process is being developed in France under the name DIAMEX and last but not least there is the TRPO method initially developed in China.

5.1. The TRUEX process

CMPO is a very powerful extractant for all III, IV and VI valent actinides from 2 to 4 M HNO_3 solutions. It is compatible with the PUREX process using TBP as extractant and displays the same radiolytic stability.

The following conclusions can be drawn with regard to the TRUEX process;

- CMPO mixed with TBP allows the extraction of the most important actinides, except Np, and the process is directly applicable to HAW solutions and to certain types of HAWC with relatively low salt content.

- The decontamination factors are of the order of 4.10^4.

- CMPO is such a powerful extractant that quantitative elution of extracted actinides, especially U and Pu, is difficult.

- CMPO is an expensive product and it needs additional purification to eliminate some interfering impurities e.g. POPPA.

- Am extracted by CMPO can be recovered quantitatively but it is accompagnied by Cm and the Rare Earths. The TRUEX process has to be complemented by an Am-Cm/RE separation (TALSPEAK) and in the end by a Am/Cm separation.

- In order to reduce the production of secondary effluents, the process ought to be modified by introducing salt free reagents (hydrazine, oxalates, hydroxylamine, ...).

- In presence of high salt concentrations third phase formation is observed.

- The TRUEX process does not generate secondary effluents but the subsequent steps (Am/Cm/RE separations) produce a lot.

5.2. DIDPA method

The DIDPA actinide separation method was developmed by JAERI as an improvement of the HDEHP method initially worked out by ANL and tested on HAW solutions by JRC Ispra. The DIDPA method makes use of Di-isodecylphosphoric acid instead of HDEHP.

- The DIDPA method needs a preliminary acid destruction step, down to 0.5 M HNO_3, prior to any actinide extraction.

- DIDPA is compatible with TBP and allows the quantitative extraction of all actinides from 0.5 M HNO_3 HAW.

- The decontamination factor is about 10^4.

- The Am-Cm-RE fraction has further to be separated with HDEHP according to the TALSPEAK process. The resulting Am-Cm fraction still contains 10 % of the accompagnying rare earths and further separation is feasible but very tricky.

- The incorporation of the Np separation into the initial process made it rather complex.

- The amount of secondary effluents is of the same order of magnitude as that of the TRUEX method.

5.3. DIAMEX process

The DIAMEX process is a dual process which relies on the one hand on the electrochemical oxydation of Am III into Am IV and on the other hand on the extractive properties of Diamides.

- Initially the mono-amides were investigated as a possible substitution of TBP. The purpose was to reduce the solid residue from solvent incineration.

- The diamides DMDOMA and DMDBMA are good extractants for all actinides from acid solutions (1-3 M HNO_3) but less powerful than CMPO or DIDPA.

- Among the diamides tested DMDBTDMA has the most attractive properties as actinide extractant in acid solution but has to be considered as a compromise between chemical properties and physical characteristics.

- The most attractive part of the present development is the selective separation of oxidized Am(IV) complexed into a phosphotungstate cage by di-n-octylamine extraction.

- The diamides are in general very viscous solvents with relatively slow kinetics and display emulsifying properties in contact with aqueous solutions.

5.4. TRPO process

This process was initially invented in the People's Republic of China and further tested at the Trans Uranium Institute at Karlsruhe.

- The TRPO extraction is similar to the TOPO extraction process which has been widely used in the mining industry during several decades.

- Though relatively new in the field of partitioning this method seems very promising because of the excellent kinetics and the complete extraction reversibility.

- The decontamination factors range from 10^3 to 10^4.

- The actinide loading capacity of TRPO is much larger than that of CMPO.

- TRPO (Tri-n-octylphosphine-oxide) is fully miscible and compatible with TBP and does not form any "third phase" with TBP-Kerosene.

- The AM-Cm-RE fraction can be selectively eluted from loaded TRPO without U-Pu cross contamination.

- Certain fission products (Zr, Mo, Ru, Tc) contaminate the actinide fraction.

- The TRPO is problably the best and most promising of the actinide partitioning methods but must be complemented as the previous methods with Am/Cm/RE sepration steps.

5.5. *Am/Cm separation*

- The largest experience has been accumulated during the production of Am and Cm isotopic heat souces. The methods developed by ORNL and SRL for the separation and purification of Am and Cm by high pressure ion exchange are undoubtedly the most extensive experimental achievement. However these methods produce huge amounts of secondary wastes.

- The most promising approach for Am/Cm/RE separation is the peroxidation to Am IV which can be followed by selective extraction, as proposed in the DIAMEX process.

6. Fuel and target fabrication

- Reprocessing of MOX fuel produces a Pu product which contains increasing amounts of Pu-238. This fact will be of utmost importance in the fuel fabrication facilities.

Table 9 gives the evolution of the Pu-238 in fresh fuel for LWR-MOX 50 Gwd/T throughout the 4 recycles and the equivalent situation for FR-MOX 129 Gwd/T.

Table 9
Evolution of the % Pu-238 in fresh Pu fuel during recycle in LWR and FR's

	R_1	R_2	R_3	R_4
LWR-MOX 50 Gwd/T 7 y discharge	3.11	3.45	3.71	3.85
FR-MOX 129 Gwd/T 17 y discharge	3.08	3.90	4.68	4.74

The increase of Pu-238 is higher in FR-MOX fuel but the amount of energy extracted from the fuel is 2.5 times higher. The fuel fabrication facilities must be desgined to treat in the future fuel types with 4 to 5 % Pu-238.

- The preparation of Am and Cm containing fuel is the greatest challenge in this field since the amounts of Cm-244 determine the total heat load and neutron dose emitted by the spent fuel. Recycling them for actinide destruction will require a fully new technologic approach which has not yet been experienced even on small scale.

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