SESSION IV

REACTOR IRRADIATIONS AND COMPARISON WITH ADS

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Six papers were presented on reactor irradiation:

- The management of Am and Cm was presented with various options – from once-through transmutation to a multiple recycling scheme. Cm transmutation greatly reduces the radiotoxicity but has substantial consequences on the fuel cycle facilities.

- On the activity of IPPE in Russia, calculational and experimental studies on MA transmutation were presented. The potential radiotoxicity reduction in the waste storage was discussed in three scenarios.

- Schemes for recycling Am targets in PWR/MOX cores were shown to be feasible, provided that enriched UO_2 is used in support of MOX fuel. Pu multiple recycling is complicated and results in a significant cost penalty.

- The neutronic performance of a small, critical, dedicated MA burner was presented. The lead-cooled, dense fuel cores are favourable for increasing the MA mass consumption rate and enable an indefinite recycling. The use of a moderator ensures an effective Doppler feedback.

- The status of fast reactor transmutation studies at JNC was presented. The future approach to FBRs and the related fuel cycle system in Japan was also introduced.

- The mass flows of Pu and MA in an LWR-FBR fuel cycle model were shown. Pu and MAs from LWRs can be consistently consumed avoiding further accumulation by gradually implementing similar scale FBRs and the new fuel cycle.

The main issue is the transmutation of Am and Cm. While the reduction in radiotoxicity due to Am and Cm transmutation is necessary, there are the problems associated with fuel fabrication and separation of Am, Cm and rare earth elements. In collaboration with experts on separation and fabrication, further investigation into recycling Am and Cm should be made to provide various options in finding the best way to transmute Am and Cm.

Three papers introduced comparisons of critical reactors with ADS:

- A comparison of neutronic characteristics and possible future R&D work for fast neutron spectrum designs of critical reactor and ADS was presented. The possibility of a thermal ADS was discussed.

- A point kinetics comparison was studied for an ADS and a traditional reactor. The ADS has a great advantage in the case of a TOP but a less favourable behaviour for a LOFWS type accident.

- The reactivity effects in molten lead energy amplifiers was investigated. To avoid reaching criticality in abnormal conditions, the normal k-eff value limitations impose restrictions on the neutron flux.

The comparison of neutronics, thermal-hydraulic characteristics, safety and cost between critical reactors and ADS should be continued in the second phase of the P&T system studies by the OECD-NEA.

TRANSMUTATION OF AMERICIUM AND CURIUM: REVIEW OF SOLUTIONS AND IMPACTS

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Abstract

Several papers have presented studies on scenarios based on Plutonium and minor actinides recycling. Recycling Neptunium allows a reduction of long-term radiotoxicity. Recycling Americium allows a reduction of radiotoxicity of more than a decade between 100 years and 10 000 years. The gain in radiotoxicity is limited due to the Curium produced in the reactors. The Curium transmutation enables greatly the radiotoxicity reduction but presents large consequences on the fuel cycle facilities. The study compares the following solutions: once-through cycle of Americium (and Curium) in dedicated assemblies in the fast reactor, dedicated fast reactor for Americium (and Curium) and the dilution of the actinides in PWR.

Introduction

Several papers [1,2] were presented on scenarios studies of actinides transmutation. This one is focused on the Americium and Curium management with various options, from a once-through transmutation to a multiple recycling scheme.

The transmutation of minor actinides (MA) is coupled on a proper use of Plutonium (Pu). The Plutonium is an energetic material and its recycling in a FBR (Fast Breeder Reactor) with the Uranium permits the use of almost all the initial Uranium and increases the energetic gain by several decades. Today, the studies for an efficient transmutation of minor actinides are linked on different ways of utilising Plutonium [in LWR, in FBuR (Fast Burner Reactor), in FBR]. Based on scenarios of Plutonium recycling, the transmutation of minor actinides is studied and different possibilities are compared.

Recycling Neptunium allows a reduction of long-term radiotoxicity (1 Million of year) and is feasible in a MOX fuel in a homogeneous mode. Several studies have already evaluated the Neptunium transmutation and will not be recalled here.

Recycling Americium allows a reduction of radiotoxicity of more than a decade between 100 years to 10 000 years, but its transmutation produces Plutonium and Curium. Thus the radiotoxicity reduction is limited to a factor 10 or less due to the Curium production in the reactors. To increase the reduction factor up to 100, it is needed to recycle the Curium.

The Curium transmutation enhances greatly the radiotoxicity reduction but presents large consequences on the fuel cycle facilities (fabrication and reprocessing plants, transport, …). In order to evaluate and compare scenarios, an evaluation of:

- The radiotoxicity in the storage.
- The mass flux per element (separated or not).
- The consequences on the cycle physic parameters (activities, neutron emission, …).

is presented in the paper for the following solutions (among others):

- Once-through of Am (and Cm) in dedicated assemblies in FR.
- Dedicated FR (CAPRA) for Am (and Cm).
- Dilution of the actinides in PWR.

Americium production within Plutonium recycling scenarios

The decrease of minor actinides inventory is based on Plutonium recycling scenarios. Indeed, Plutonium is the major element for radiotoxicity and mass in the storage. Table 1 describes the minor actinides production in nuclear park with Plutonium recycling in PWR (Pressurise water reactors) or FR (Fast reactors).

Table 1. **Mass flow to the storage kg/TWhe for a nuclear park (400 TWhe, 60 GWe)**
(Loss: 0.1% for Pu)

	100% PWR UOX (open cycle)	100% FR	32% FR + PWR UOX	18% FR + PWR UOX & MOX	100% FR MOX with enriched U
Pu	28.1	0.16	0.09	0.08	0.05
Np	1.7	0.50	1.2	1.3	1.5
Am	1.3	3.2	4.3	4.6	4.5
Cm	0.26	0.34	0.75	1.0	2.25

The consequences are calculated on the radiotoxicity. The gain on this parameter with Plutonium recycling compared to an open cycle is between a factor 3 and 6 for the time scale (100/10 000 years). The radiotoxicity is then dominated by the Americium, and after, the Curium, the Plutonium losses and the Neptunium for the same time scale. The Curium masses produced are significant, 10% to 20% of the amercium masses for a Plutonium recycling and up to 50% for the Plutonium recycling in a MOX with enriched Uranium support. This last concept, allowing the feasibility, has lower performances compared to a 100% FR park (50% for the Americium and a factor 7 for the Curium).

The once-through option for Americium and Curium recycling

The once-through recycling with a high fission rate (more than 90 % of atoms) is a very attractive solution because:

- The production of Curium due to Americium recycling is managed inside the concept.
- The reprocessing of the target can be simplified, compared to multiple reprocessing (just clean the material, if needed).

The gain on the radiotoxicity varius the fission rate in the target, compared to the open cycle are given in Table 2 for the nuclear park and given in the 4th column of Table 1 (PWR UOX§MOX and FR).

Table 2. **Reduction of radiotoxicity compared to open cycle, using targets in dedicated assemblies (moderated) in FR**

Factor of the radiotoxicity reduction/open cycle	100 years	1 000 years	10 000 years
Open cycle	1	1	1
Pu recycling	3	2,5	4.5
recycling Pu + targets Am			
Maximum (Fission rate 100%)	20	23	15
Fission rate = 90%	12	17	10
Fission rate = 95%	16	20	13
Fission rate = 98%	17	21	14
Pu recycling + targets Am + Cm			
Maximum (Fission rate 100%)	490	400	390
Fission rate = 90%	40	45	30
Fission rate = 95%	72	81	55

For an Americium recycling (without Curium recycling), the maximum gain reachable at 10 000 years after irradiation is clearly limited by the Curium provided by the nuclear park (in the PWR UOX § MOX and in FR). The value at this time is the minimum gain for the time scale between 100 years and 1 million of years. This is why we choose it as a key parameter to compare the transmutation impact in this study.

In order to have an equilibrium between the production of Americium in the park and the transmutation in the dedicated assemblies disposed in FR with a fission rate of 90%, we need slightly more than 30% of FR in the nuclear park.

In this scenario, the gain of radiotoxicity is about 10 compared to an open cycle (see table 2). This gain is small.

More the FR fraction is large, more the radiotoxicity reduction at 10 000 years will be close to the maximum value (100% for the fission rate) due to the smaller build-up of Americium and the larger efficiency in the targets (larger moderation with high flux). Anyway, the gain on the radiotoxicity is limited to a factor 10 to 20 compared to an open cycle. To go further, it is needed to have a Curium and Americium recycling with a fission rate in the targets of about 95% or more. In this case, it is difficult to obtain an equilibrium between transmutation and production in the park if we do not have a majority of FR.

Multiple recycling of Americium and Curium

An other way than transmutation in a once-through mode is multiple recycling in a homogeneous way in FR in dedicated reactors [3]. The radiotoxicity reductions are linked to the losses during the reprocessing.

The gains in radiotoxicity in this case are given in the Table 3.

Table 3. **Reduction of radiotoxicity compared to open cycle**

Factor of the radiotoxicity reduction open cycle	100 years	1 000 years	10 000 years
Open cycle	1	1	1
Pu recycling	3	3	4.5
Pu + Am			
Maximum	20	23	15
Homogeneous recycling Pu, Am (losses *=1%)	7	10	6
Homogeneous recycling Pu, Am (losses *=0.1%)	8	10	6.5
Pu + Am + Cm			
Maximum	490	400	390
Homogeneous recycling Pu, AM (losses***** =1%)	90	100	120
Homogeneous recycling Pu, AM (losses***** =0.1%)	270	335	310
(if losses (Pu, Am, Cm) = 0.01 %)	1000 <	1000 <	1000 <

* **losses:** loss fraction of A.M – For Pu: 0.1 % in all cases.

The transmutation of Americium and Curium are more favourable in a multiple recycling than in a once-through cycle, the multiple recycling being closer to the maximum values (total transmutation in fission products). The gains are sensitive to the losses rates.

Once through recycling - multiple recycling

The previous results show the interest of a Curium and Americium recycling. The consequences on Curium recycling on the cycle facilities must be evaluated.

For the fabrication

In a once-through scenario, added 10 to 20% of Curium in a fresh fuel to the Americium leads to an increase of power, γ doses and a large gap in the neutron emission. Table 4 gives the main effects when Curium is added to Americium in a target.

Table 4. **Impact of the recycling of Americium and Curium for the fabrication of a target (mass ratio ≅ 20 % of M.A. on inert support)**

	100 % Am	90 % Am + 10 % Cm	80 % Am + 20 % Cm
Power	1	× 2.3	× 3.6
G doses at 1 m	1	× 1.5	× 2
Neutrons source	1	× 120	× 240

In a multiple recycling scenario, impacts are lower. The multiple recycling of Americium and Curium presents lower impacts than for the Curium recycling compared to Americium recycling in a heterogeneous disposition.

Table 5. **Impact of the recycling of Americium and Curium at fabrication**

	Heterogeneous recycling (Target ≅20%)		Homogeneous recycling FR (2.5%)	
	Am	Am+Cm	Am	Am+Cm
Power	× 14	× 32	× 1.7	× 3.9
Dose ϒ	× 1 700	× 2 500	× 76	× 115
Neutron source	× 7	× 840	× 1.4	× 170

Ref: Standard fuel EFR CD 9/91.

The dilution allows to reduce the impact.

For reprocessing

The impact on reprocessing is limited, even for the neutron source, as is shown in Table 6.

Table 6. **Impact of the recycling of Americium and Curium**

	Heterogeneous recycling (Target ≅20%)		Homogeneous recycling FR (2.5%)	
	Am (FR) 90%	Am+Cm (FR) 90%	Am	Am+Cm
Power	× 32	–	× 3	× 3.3
Dose ϒ	÷3.5	–	× 1	× 1
Neutron source	–	–	× 3.5	× 4

Ref: Standard fuel EFR CD 9/91.

The tables show a maximum increase of factor 5 at reprocessing step for the option of homogeneous recycling, and larger for heterogeneous recycling.

Once-through or multiple recycling?

The option of Americium and Curium multiple recycling is still open. This solution allows larger radiotoxicity gains and the homogeneous mode is more efficient than once-through. We can optimise once-through and multiple recycling by a two step once-though, limiting the reprocessing mass flux.

This analysis shows the interest of reprocessing with a high resistance to the neutron emission and to the activity in order to reach a gain on radiotoxicity larger than 100.

Conclusions

- The Americium recycling alone allows small gains on radiotoxicity at 10 000 years.
- To increase this gain, the Curium recycling is needed with the Americium.
- The need of a reprocessing with strong resistance is underlined by these studies.
- This is the same for the fabrication route and fuel stability.
- The main options with a large gain in radiotoxicity and a first feasibility assessment in the reactor are:
  - once-through recycling in dedicated assemblies in FR, in a large number of FR.
  - multiple recycling: in PWR, in all the reactors; in FR, with the Plutonium and in a large number of FR.

REFERENCES

[1] M. Delpech *et al.,* GLOBAL ’97, *Scenarios of Plutonium and Minor Actinide Recycling*, Mito, Japan.

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[3] J. Tommasi *et al*., GLOBAL ’97, *Heterogeneous Recycling of Americium in Thermal and Fast Reactors*, Mito, Japan.

CALCULATION AND EXPERIMENTAL STUDIES ON MINOR ACTINIDES REACTOR TRANSMUTATION

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Abstract

Brief survey of the activity being carried out in IPPE in the field of calculation and experimental studies on MA transmutation is given. Results of recent developments on calculation code and cross-sections supply for MA transmutation works are presented. Calculation investigations on incineration of Am and Cm with shares of rare-earth elements in BOR-60 reactor are described for the forthcoming experiment. Comparative analysis on MA transmutation in fast and thermal reactors is given.

Introduction

Nowadays in Russia the program on transmutation of MA and long-lived fission products is developed only on each next year (due to lack of financing supplied by the Government for development of the whole nuclear power). Therefore, the schedule of works is corrected each year to provide fulfilment of the most valuable tasks. Nevertheless, the large volume of works has been done, in particular, thanks to the assistance of our foreign colleagues supporting realisation of our plans in the joint projects.

General review

As it was in previous years, the main directions of the activity in the field of MA and long-slived fission products transmutation in Russia are as follows:

1. Modernisation of nuclear constants data and software codes needed for calculation studies on MA and long-lived fission products transmutation.
2. Development and calculation-experimental grounding of the specialised fast reactor cores for incineration of Plutonium, MA and some fission products.
3. Investigation of closed fuel cycle with Plutonium and MA recycling.
4. Carrying out integral experiments on the critical assemblies simulating power reactors to correct the data on MA nuclear reactions cross-sections.
5. Preparation and carrying out experiments in power reactors on MA sample fuel irradiation.

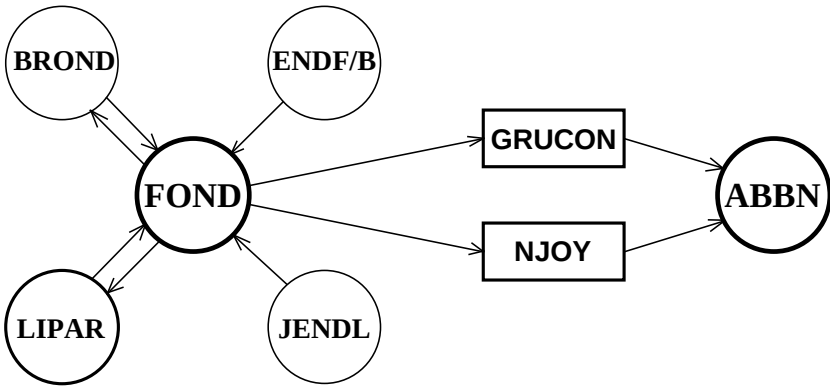
Nuclear constants data and software for minor actinides transmutation calculations

Calculations of MA transmutation in fast reactors base on the TRIGEX [1] code performing diffusion 3D reactor calculation in HEX-Z geometry with ABBN-93 [2] nuclear constants. Software system CONSYST2/ABBN (see DLC-182/ABBN-90 in the RSICC Data Library Collection) provides ABBN-93 constants binding with TRIGEX code by means of PRECON1 subroutine. TRIGEX code performs calculations of fuel composition variation with burn-up using CARE [3] code.

Calculations of MA transmutation in thermal reactors at present are carried out by means of Monte-Carlo method codes MCU-RFFI (RRC KI, Moscow), KENO (being the part of the American calculation complex SCALE 4.3 [4]) and MCNP. For cells and subassemblies calculations, TBC-M code (RRC KI, Moscow) is used. Constants preparation for the KENO code is realised through WIMS-ABBN code as well as by CONSYST code being the part of the CONSYST2/ABBN nuclear constant supply system with ABBN-93 nuclear constants. Constants binding is performed using ANISN-like formats. CONSYST2/ABBN system application supports carrying out of calculations performed by both precise codes implementing Monte-Carlo method, and kinetic transport reactor calculations in DSN-PN approximation, such as ANISN, TWODANT and DOT.

ABBN-93 constant system is the most modern version of the ABBN group constants. It is created on the base of evaluated neutron data library files FOND-2 [5], which integrates files from Russian library BROND-2, foreign libraries ENDF/B-VI, JENDL-3, and some other sources (Figure 1).

Figure 1. **ABBN-93 constact system**



Calculation of the nuclide transformation chains during MA transmutation calculations is performed by both CARE code and ORIGEN code which is the component of the American calculation complex SCALE 4.3.

Calculating the nuclide transformation, resonance shielding of neutron cross-sections is taken into account for the isotopes being members of the media composition (from TRIGEX code). For all the rest isotopes needed for transformation chains calculation, cross-section data is being taken from the ABBN external libraries of the group neutron cross-sections: FP - library of fission product nuclei, ACT - library of actinides. Library of masses is also used additionally.

Multi-group library of fission product nuclei FP contains data on the radiation capture cross-sections for 169 nuclides:

^{75}As, ^{76}Ge, ^{77,78,79,80,82}Se, ^{81,82}Br, ^{82,83,84,85,86}Kr, ^{85,86,87}Rb, ^{86,88,89,90}Sr, ^{89,90,91}Y, ^{90,91,92,93,94,95,96}Zr, ^{95}Nb, ^{95,96,97,98,99,100}Mo, ^{99}Tc, ^{100,101,102,103,104,106}Ru, ^{103,105}Rh, ^{104,105,106,107,108,110}Pd, ^{109,110m,111}Ag, ^{100,110,112,113,114,115m,115n,116}Cd, ^{115}In, ^{115,116,117,118,119,120,121,122,123,124,125,126}Sn, ^{121,122,123,124,125,126,127}Sb, ^{122,123,124,125,126,127m,128,129m,130,132}Te, ^{127,129,130,131}I, ^{128,129,129m,130,131,131m,132,133,133m,134,135,136}Xe, ^{133,134,135,136,137}Cs, ^{134,135,136,137,138,139,140}Ba, ^{139,140}La, ^{140,141,142,143,144}Ce, ^{141,143}Pr, ^{142,143,144,145,146,147,148,150}Nd, ^{147,148,148m,149}Pm, ^{147,148,149,150,151,152,153,154}Sm, ^{153,154,155,156}Eu, ^{154,155,156,157,158,160}Gd, ^{159,160,161}Tb, ^{160,161,162,163,164}Dy.

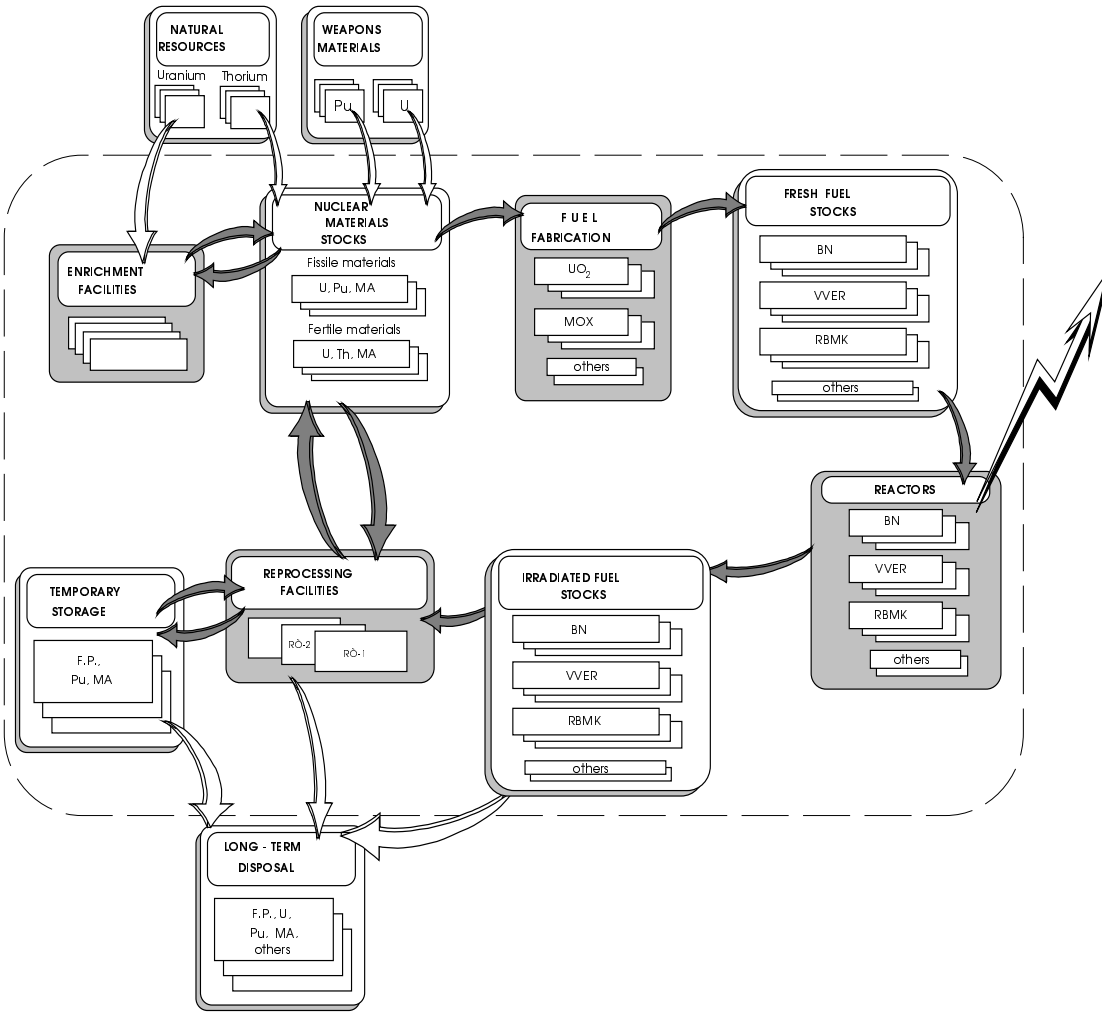
Actinides library ACT contains group cross-sections of (n,2n), (n,3n), (n,γ) and (n,f) reactions for the following nuclei:

^{223,224,225,226}Ra, ^{225,226,227}Ac, ^{227,228,229,230,232,233,234}Th, ^{231,232,233}Pa, ^{232,233,234,235,236,237,238}U, ^{236,237,238,239}Np, ^{236,238,239,240,241,242}Pu, ^{241,242,242m,243,244,244m}Am, ^{241,242,243,244,245,246,247,248,249,250}Cm, ^{249,250}Bk, ^{249,250,251,252,254}Cf, ^{254,255}Es, ^{255}Fm.

For the nuclear fuel cycle simulation, CANFU [6] code is developed. The main purpose of the CANFU code is concluded in automation of physical, radiation and ecological characteristics of the nuclear fuel cycle. To perform the above mentioned task, the code is designed and developed as a kind of meccano, allowing to create and arrange various schemes of a fuel cycle using predefined fuel cycle components (reactors, fuel management facilities, storages, etc.) with standard or user-defined parameters. The code takes use of the program modules for physical characteristics calculation (TRIGEX, CARE) and nuclear data library (ABBN-93). Figure 3 represents sample scheme of nuclear fuel cycle being simulated in the CANFU code.

Application of CANFU code allows to perform calculation studies of closed fuel cycles with both Plutonium and minor actinides recycling.

Figure 2. **Sample fuel cycle model implemented in CANFU code**



Development of specialised fast reactors for Plutonium and minor actinides incineration

Fast reactors are capable not only to effectively incinerate Plutonium of any isotopic composition, they can be used also for efficient utilisation of minor actinides. The main physical problem being arisen is connected with the degradation of void reactivity effect. However, cores with increased fuel enrichment have some resource in SVRE value, allowing to introduce (homogeneously) up to 5-7% of minor actinides in fuel. Transmutation efficiency in this case can reach up to 50 kg annually.

As the most effective incinerators of both Plutonium and minor actinides, cores should be considered with the fuel not containing ^{238}U. The main tasks to be solved on the way of such cores creation are increase of safety due to Doppler-effect growth and fuel reloading schedule optimisation. Numerous investigations carried out recently in IPPE permitted to optimise such a core using new

fuel material (PuO_2+MgO+Fe with MA addition up to 15% vol.), which has been also certified. Utilisation of such kind of fuel allows to increase MA incineration efficiency up to 100-150 kg annually. Table 1 presents calculated values of MA incineration efficiency [7] for the various cores of fast reactor with capacity 800 MWt.

Table 1. **Comparison of calculated values of MA incineration efficiency for the various cores**

	Cores with the MOX fuel of increased enrichment			Cores with the fuel not containing ^{238}U		
MA Location	Homogeneously in the fuel	Heterogeneously in the core	In the radial blanket	Homogeneously in the fuel		In the radial blanket
Kind of fuel with MA	MOX	MOX	MAO_2 in inert matrix	PuO_2 +MAO_2	$^{235}UO_2$+MAO_2	MAO_2 in inert matrix
Efficiency of minor actinides incineration, kg/year	**50**	**50**	**50**	**120**	**260**	**80**

Further increase of MA incineration efficiency is possible with substitution of Plutonium as a fuel material by ^{235}U.

Experimental studies

Estimations of requirements have been performed in SSC RF IPPE on actinides neutron cross-sections correction for the calculation of recycling parameters for the purpose of transmutation. The estimates show that with the increase of actinides share in the core, contribution to the whole K_{eff} uncertainty is stipulated for the uncertainties in fission and elastic scattering cross-sections of ^{237}Np, ^{241}Am, ^{243}Am isotopes. With the share of actinides in fuel equal to 25%, the K_{eff} uncertainty makes up 2.6%, and due to correction of actinides neutron cross-sections it can be reduced up to 1.5%. In this connection, carrying out of experiments on BFS facilities and power reactors is being continued.

During several years in SSC RF IPPE experimental program is being intensively realised aimed at detailisation of data on MA in integral experiments on BFS critical facilities. Experiments of various types have been carried out:

- measurement of fission cross-section ratios of MA isotopes (there are fission chambers with layers of Am, Np, Cm, Pu, Th, U on the stands) to the standard ones (^{239}Pu or ^{235}U) on the series of fast critical assemblies with various neutron spectrums;
- measurement of central reactivity worths of MA sample, which gives an opportunity to obtain data on capture cross-sections taking into account previously mentioned information;
- estimation of the influence of introduction of considerable quantity of ^{237}Np in fuel on the main neutron-physical characteristics of cores (void effect of coolant, Doppler-effect, spectral indices, etc.).

To carry out the experiments with introduction of Neptunium into the core, 250 BFS pellets (about 11 kg) with Neptunium dioxide have been produced. Then the blocks with Neptunium dioxide have been placed in central parts of the cores of Superphenix and CAPRA type, simulating 13% and 6.5% Neptunium content in fuel.

According to the program being planned of the MOX fuel studying in light-water reactors on SUPR facility being under construction, investigations are intended on MA burning in thermal spectra as well.

The series of experiments is being completed on irradiation of actinide samples in BN-350 reactor. At present the samples of ^{238}Pu, ^{237}Np, ^{241}Am, ^{243}Am irradiated in 80-90th are delivered to the hot cell in IPPE.

Preparation of works in on the way on irradiation of experimental AMOX fuel elements in BOR-60 reactor. Calculations of isotopic composition evolution and decay heat of AMOX fuel have been performed as a part of works on the branch program “RECYCLE”.

The calculations took into account the following assumptions on the composition of AMOX fuel for BOR-60 reactor:

1. the basis of the fuel consists of Uranium with the enrichment 75%.
2. Americium-Curium fraction obtained during reprocessing of VVER-1000 reactor spent fuel with starting enrichment 4.4% and burnup 40 GWt d/t is then added in proportion 20% or 50% mass.
3. this fraction, in its turn, contains impurity of 10% rare-earth elements not separated from the Americium and Curium during chemical reprocessing.
4. The chemical form of actinides in fresh fuel has been adopted as follows: Uranium - UO_2, Americium and Curium - Am_2O_3 and Cm_2O_3.

Calculated values of the isotopic composition of source Americium-Curium mixture is given in Table 2.

Table 2. **Isotopic composition of Am-Cm fraction in VVER-1000 reactor spent fuel with 10 year cooling.**

Isotope	Content, %
^{241}Am	83.746
^{242}Am	1.22E-06
^{242m}Am	0.101
^{243}Am	13.163
^{242}Cm	2.46E-04
^{243}Cm	0.029
^{244}Cm	2.731
^{245}Cm	0.227
^{246}Cm	3.59E-03
^{247}Cm	6.31E-05
^{248}Cm	6.25E-06

Some important numerical results are presented in Table 3.

Table 3. **The most important characteristics of the transmutation process of Americium-Curium mixture being a part of AMOX fuel of BOR-60 reactor.**

Parameter	20% Am-Cm	50% Am-Cm
Transmutation of Americium, g/t h.a.	32500	81300
Build-up of Curium, g/t h.a.	4448	10940
Build-up of Plutonium, g/t h.a.	14700	28370
"Incineration of Americium" (mass of transmutated Americium excluding sum mass of Plutonium and Curium built-up), g/t h.a.	13352	41630
Build-up of helium, g/t h.a.	151	378
Specific Am transmutation, g/GWt hour (t)	10500	30100
Specific Am incineration, g/GWt hour (t)	4300	15400

In course of transmutation of Americium added to the BOR-60 conventional fuel during the refuelling interval about 21% ^{241}Am and 13% ^{243}Am is transmuted; accumulation of Curium isotopes in spent fuel increases in thousand times as compared to the initial conventional fuel; accumulation of ^{238}Pu and ^{242}Pu increases in dozen and hundred times; mass of helium released under irradiation increases in hundred times, during the storing period mass of ^{237}Np raises in dozen times as compared to MOX fuel.

The main feature of spent AMOX fuel is extremely high decay energy release and intensity of neutron emission, in comparison with Uranium and even MOX fuel. The absolute value of the sum decay energy release remains at the level exceeding 1 MWt/t during approximately 1 month. This circumstance is of importance with respect to one of the main problems of reactor safety - cooldown at the emergency shutdown with heat removal cessation - and should be studied in detail.

Some results on actinide balance in various scenarios of Pu and MA Recycling

The present chapter presents calculation studies of physical characteristics of fuel cycles based on thermal reactors exclusively as well as of fuel cycles with thermal reactors and specialised fast reactors with high enrichment on Plutonium. Material indices on Plutonium consumption and radioactive waste accumulation have been calculated. Equilibrium isotopic compositions of fuel being reached in closed systems of reactors have been estimated. Basing on the same power production level, waste radiotoxicity values are given for the scenarios considering different types of reactors. The main goal of the work was to show how the accumulation of MA changes in the wastes of fuel cycle with introduction of specialised fast reactor.

Description of the considered scenarios

Three different scenarios of fuel cycle have been considered:

1. Open fuel cycle with VVER-1000 type reactors operating on civil Plutonium.
2. Closed fuel cycle with Pu recycling in VVER-1000 reactor.
3. Closed fuel cycle with Pu recycling in VVER-1000 reactor and MA recycling in specialised fast reactor FR. with capacity 1300 MWt for MA incineration.

Calculations of long-lived radioactive wastes accumulation have been performed under condition of equal power production level (100 GWt(e) year) for each scenario.

In the open fuel cycle at the beginning of the irradiation stage fresh fuel had the same isotopic composition, and after irradiation all the spent fuel has been transported to waste storing facility. During the period of reactor operation, isotopic composition of heavy radioactive nuclei in the storage change due to radioactive decay and receipt of next portion of spent fuel from cycle to cycle.

In the calculation model of the closed fuel cycle, irradiated fuel of VVER-1000 reactor has been reprocessed extracting Plutonium and Uranium, which has been used for the production of fuel for the next cycle. The wastes in this case were formed by fission products and MA from the spent fuel. The new fuel has been enriched by the initial civil Plutonium, diluted by depleted Uranium and sent again to the irradiation in reactor. For the FR reactor, all heavy isotopes have been extracted from the spent fuel, MA from VVER spent fuel have been added and after enrichment this new fuel has been sent to the next cycle of irradiation. The extraction extent for all heavy isotopes during chemical reprocessing has been adopted as 0.999.

Isotopic composition of the feed Plutonium for all scenarios was equal to the isotopic composition of civil Plutonium (%):

$$^{238}Pu/\ ^{239}Pu/\ ^{240}Pu/\ ^{241}Pu/\ ^{242}Pu = 0.9/61.0/22.0/12.0/4.1$$

The main characteristics of VVER-1000 and FR reactors operating in open fuel cycle are presented in Table 4.

Table 4 **The main characteristics of VVER-1000 and FR reactors**

Reactor Parameter	VVER-1000	FR
Electric power (MWt)	1000	1300
Type of fuel	MOX	MOX
Number of subassemblies in core	163	349
Enrichment, %	6.6	25.3/29.34
Burnup, GWt days/t	40	70
Irradiation interval, eff.days	876	1200
Number of reloads	3	3
Load factor	0.8	0.8
Start loading, t (h.a.)	65.25	34.51
Loading, t/year(h.a.)	21.75	8.4
Pu loading, kg/year	1435.5	2277.5
Pu discharge, kg/year	1040.5	1840.6
Pu balance, kg/year	-395	432
Pu isotopic composition in spent fuel (%)		
^{238}Pu	1.84	0.94
^{239}Pu	42.55	56.01
^{240}Pu	28.05	29.79
^{241}Pu	18.76	7.90
^{242}Pu	8.79	5.36

Calculation results

Figures 3, 4, 5 illustrate the balance of fuel nuclides for 3 different scenarios under the same power production rate 100 GWt·year.

Figure 3. **The balance of fuel nuclides in open fuel cycle (Scenario 1)**

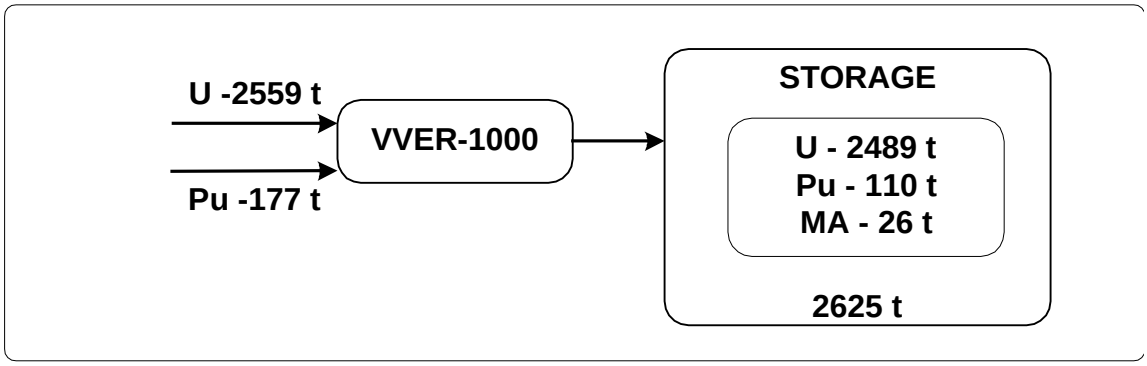


Figure 4. **The balance of fuel nuclides in closed fuel cycle with U and Pu recycling in VVER reactor (Scenario 2)**

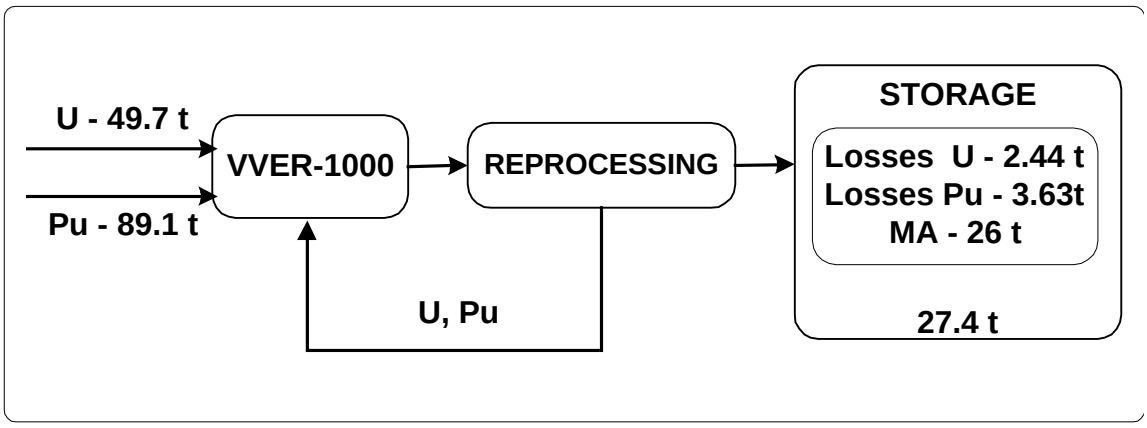
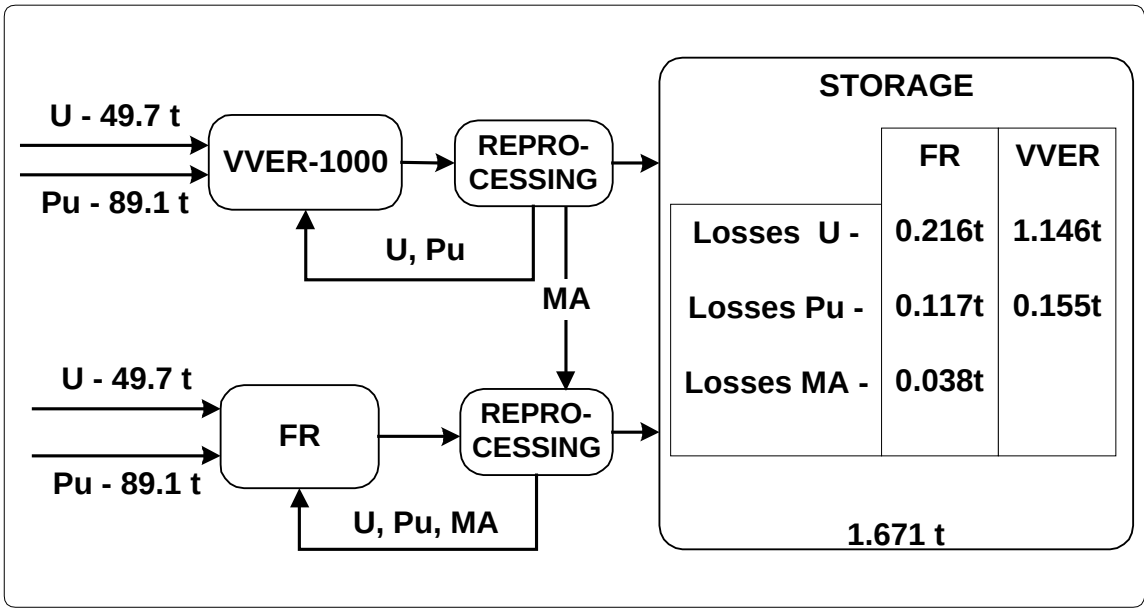


Figure 5. **The balance of fuel nuclides in closed fuel cycle with U and Pu recycling in VVER reactor and U, Pu, MA recycling in FR reactor (Scenario 3)**



In course of recycling equilibrium compositions of loaded and discharged fuel are set. Tables 5 and 6 show the compositions of loaded and discharged fuel for VVER-1000 and FR reactors. It can be seen from the tables that in the system consisting of VVER-FR reactors 4 kg of Np, 207 kg of Am and 47 kg of Cm are transmuted per year.

Table 5. **Equilibrium compositions of loaded and discharged fuel set in VVER-1000 reactor in course of Pu recycling**

	Loading, kg/cycle	Discharge, kg/cycle	Balance, kg/cycle	Balance, kg/year
^{235}U	31	28	2	1
^{238}U	57941	56753	1188	495
^{237}Np	0	10	-10	-4
^{238}Pu	254	235	19	8
^{239}Pu	2884	1561	1323	551
^{240}Pu	1805	1329	477	199
^{241}Pu	1147	887	260	108
^{242}Pu	1121	1033	88	37
^{241}Am	0	230	-230	-96
^{242m}Am	0	3	-3	-1
^{243}Am	0	267	-267	-111
^{242}Cm	0	0	0	0
^{243}Cm	0	1	-1	0
^{244}Cm	0	103	-103	-43
^{245}Cm	0	10	-10	-4
^{246}Cm	0	0	0	0

Table 6. **Equilibrium compositions of loaded and discharged fuel set in FR reactor in course of Pu and MA recycling**

	Loading, kg/cycle	Discharge, kg/cycle	Balance, kg/cycle	Balance, kg/year
^{235}U	42	39	3	1
^{238}U	17108	15460	1647	501
^{237}Np	71	58	13	4
^{238}Pu	727	709	18	5
^{239}Pu	4722	3460	1262	384
^{240}Pu	5248	4795	453	138
^{241}Pu	982	732	250	76
^{242}Pu	1292	1207	84	26
^{241}Am	1183	870	313	96
^{242m}Am	61	57	3	1
^{243}Am	1510	1145	364	111
^{242}Cm	1	1	0	0
^{243}Cm	10	9	1	0
^{244}Cm	1059	918	140	43
^{245}Cm	219	205	14	4
^{246}Cm	100	100	0	0

For the Scenario 3, equilibrium compositions of Pu being loaded in VVER reactor and MA being loaded in FR reactor are of interest. Figure 6 indicates variation of Pu isotopic composition in the loading of VVER-1000 reactor. Fuel enrichment in this case increases from 6.6% up to 10%. Figure 7 illustrates variation of mass of actinides being loaded in FR reactor in recycling of MA built-up by both FR and VVER reactors. Fuel enrichment of FR reactor in equilibrium increases up to 34% and 40% for inner and outer cores accordingly.

Figure 6. **Variation of Pu isotopic composition in the loading in course of its recycling in VVER-1000 reactor**

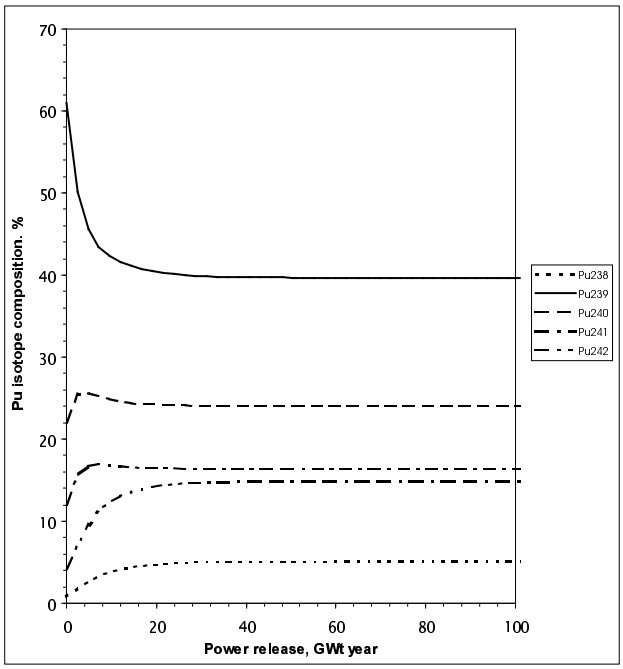


Figure 7. **Variation of MA content in the loading in course of its recycling in FR reactor**

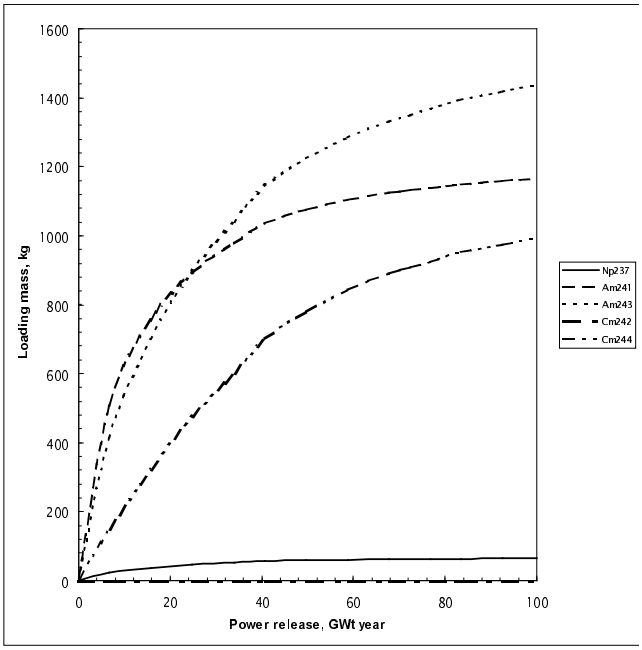
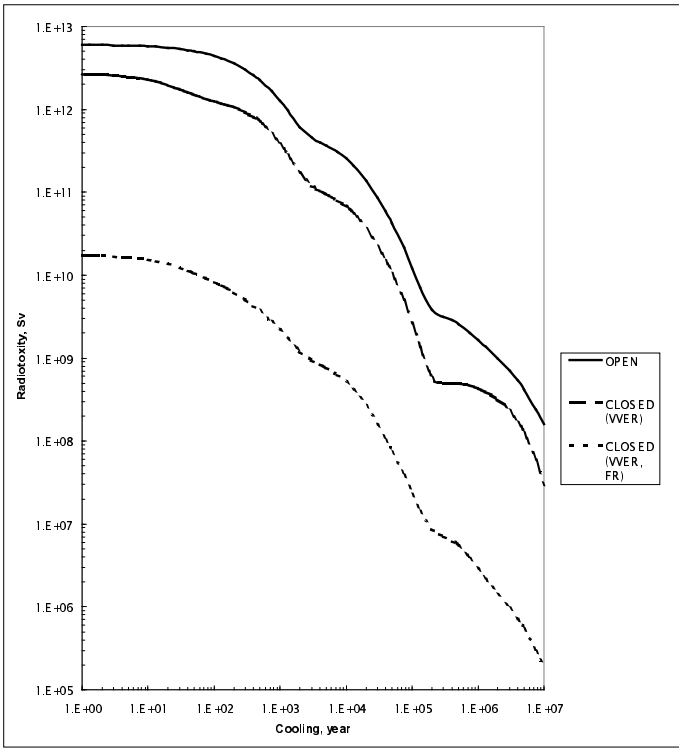


Figure 8 presents potential sources of radiotoxicity of long-lived nuclides formed in storage for three scenarios under consideration. The curves are obtained by means of simple multiplication of activity of radionuclides formed in storage during cooling by their hazard factor. It follows from the figure that closure of fuel cycle only on Plutonium and Uranium does not affect dramatically on the change of potential radiotoxicity. Decrease of radiotoxicity in this case constitutes merely 2-3 times. Recycling of all actinides, as it can be seen from the curve corresponding to Scenario 3, decreases radiotoxicity in 100-150 times.

Figure 8. **The sources of potential radiotoxicity in the waste storage for three considered scenarios**



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RECYCLING SCHEMES OF AMERICIUM TARGETS IN PWR/MOX CORES

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Abstract

From the orientation studies performed so far, both ways to recycle Am in PWR/MOX cores, homogeneous in MOX or heterogeneous in target pins, appear feasible, provided that enriched UO_2 is used as support of the MOX fuel. Multiple recycling can then proceed and stabilise Pu and Am quantities.

With respect to the Pu multiple recycling strategy, recycling Am in addition needs 1/3 more ^{235}U, and creates 3 times more Curium. Thus, although feasible, such a fuel cycle is complicated and brings about a significant cost penalty, not quantified yet.

The advantage of the heterogeneous option is to allow to manage in different ways the Pu in MOX fuel and the Am in target pins. For example, should Am remain combined to Cm after reprocessing, the recycling of a mix of Am+Cm could be deferred to let Cm transform into Pu before irradiation. The Am+Cm targets could also stay longer in the reactor, so as to avoid further reprocessing if possible.

Introduction

Fast neutron reactors are best suited to burn Plutonium and, if required, Americium. But in case the deployment of these reactors comes to be largely delayed, it is useful to investigate under which conditions Americium could be recycled in PWR cores, provided it can be recovered at fuel reprocessing.

Americium recycling could proceed either in a homogeneous way if a mixed oxide (U, Pu, Am)O_2 is manufactured and irradiated; or in a heterogeneous way if special Am target pins are fabricated and irradiated together with the usual (U, Pu)O_2 MOX fuel.

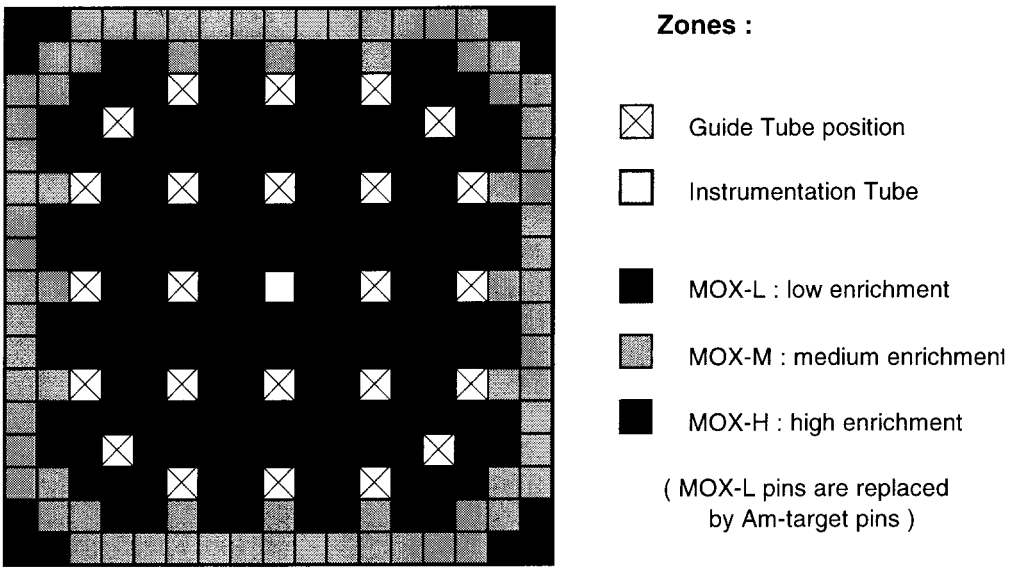
These two recycling modes have been modelled in detailed PWR core physics calculations. Results of homogeneous recycling were reported earlier [1] and are only briefly recalled here. The results of heterogeneous recycling calculations are discussed in details. The problems associated with fuel and target manufacture, will shortly be recalled, too.

Basic assumptions and methods of calculation

A nuclear network made of 900-MWe PWRs is taken as a basis, with a quarter-core MOX fuel management and an average discharge burn-up of 44 GWd/t. To allow an equilibrium to be reached between transUranium production and consumption, the Plutonium and Americium quantities recovered from reprocessing, 5 years after PWR core discharge, are refabricated over 2 years and then recycled in the same type of reactor. This recycling has been simulated explicitly, using a macrocell comprising a MOX assembly and its neighbouring UO_2 assemblies, a number of successive times large enough to verify that equilibrium is effectively reached.

Figure 1 shows the sketch of the MOX assembly, subdivided into three concentric zones of different Plutonium enrichments, so as to flatten the power and burn-up distribution in the radial direction.

Figure 1. **Sketch of MOX Assembly**



The MOX fuel is designed to be equivalent in energy to UO_2 fuel enriched to 3.7 % in ^{235}U. At discharge all assemblies, loaded with UO_2 or with MOX, will have been irradiated 4 calendar years at a load factor of 75 %.

Core physics calculations were run using a recent version of the WIMS code package [2] and a JEF 2.2 based library with 172 neutron energy groups. This library is well adapted to high-burn-up MOX fuel irradiation: as an example, the giant low-energy resonance of ^{242}Pu at 2.7 eV is covered by 8 groups. Also, the burn-up chains explicitly include 3 Americium isotopes and 4 Curium isotopes.

Homogeneous recycling of Plutonium and Americium

In the case of homogeneous recycling [1], the MOX fuel pins contain, either (UO_2PuO_2) as usual, so as to provide a reference, or ($UO_2PuO_2AmO_2$) in the variant MOX-X. First of all, calculations were done using depleted UO_2, containing only 0.25% ^{235}U, as MOX fuel support.

For the MOX-X recycling case, the required Pu+Am enrichments grow significantly with the successive recycling steps. In the reference MOX (Pu) case, these are calculated to be 7.4, 9.1 and 10.4%, while in the MOX-X case they would reach 8.7, 12.2 and 15.1 %, respectively. This leads to serious difficulties to keep the MOX loading compatible to the usual design and safety objectives. It is generally admitted that a Pu enrichment of 12 % is about the upper limit to keep non-positive coolant void reactivities. This void effect has been calculated for the beginning-of-life situations with fresh fuel which are most penalising. Only the MOX-X1 step indeed retains a non-positive void effect; the subsequent steps MOX-X2 and MOX-X3 would have positive void values, what is not presently acceptable.

Two remedies were proposed in [3] to keep Pu enrichments in an acceptable range: enhanced moderation, or use of enriched Uranium as MOX support.

The enhanced moderation consists of increasing the pin spacing (or reducing the pin thickness) so that the MOX assembly gets a larger water moderation. With the standard PWR lattice the volume ratio of water to oxide is 1.9; one could increase it to 3 or more (but the core volume is also increased). To identify the trends, calculations have been run for a few recycling steps in the same 900-MWe PWR conditions as above.

Thanks to the enhanced moderation, the necessary Pu enrichments are substantially lowered. When recycling Pu only, they become, respectively 4.75% (MOX1) and 6.3% (MOX2), instead of 7.4 and 9.1%. When recycling Pu+Am, they become 5.25% (MOX-X1) and 8.0% (MOX-X2).

This fuel concept can very effectively burn ^{239}Pu, but it also produces much ^{242}Pu, source of ^{243}Am. While this helps to stabilise total Pu quantities at a lower level, the Am quantities are not efficiently decreased, and the Cm quantities are growing.

The enhanced moderation, very promising in the first steps, is however not a better solution with repeated recycling as the enrichments again become prohibitive and the void effect positive. Such conclusions meet those found in [4]: at equilibrium the Pu enrichment reaches about 18%, which is by far excessive.

Another remedy was thus tried, which is to replace in the MOX fuel the support Uranium, usually depleted, by Uranium enriched in ^{235}U. So the Pu enrichment of the MOX fuel can be kept

equal to a constant, acceptable value, for example 8% in the average. UO_2 with an increasing enrichment would thus be added to PuO_2 as multiple recycling proceeds, so as to compensate for the progressive increase of the Plutonium quantities to be recycled and their isotopic degradation. In such a way, an equilibrium can indeed be reached where the discharged Plutonium remains identical to the loaded one, in quantity and quality. The same approach can be applied as well to the combined recycling of Pu+Am, recovered at each step.

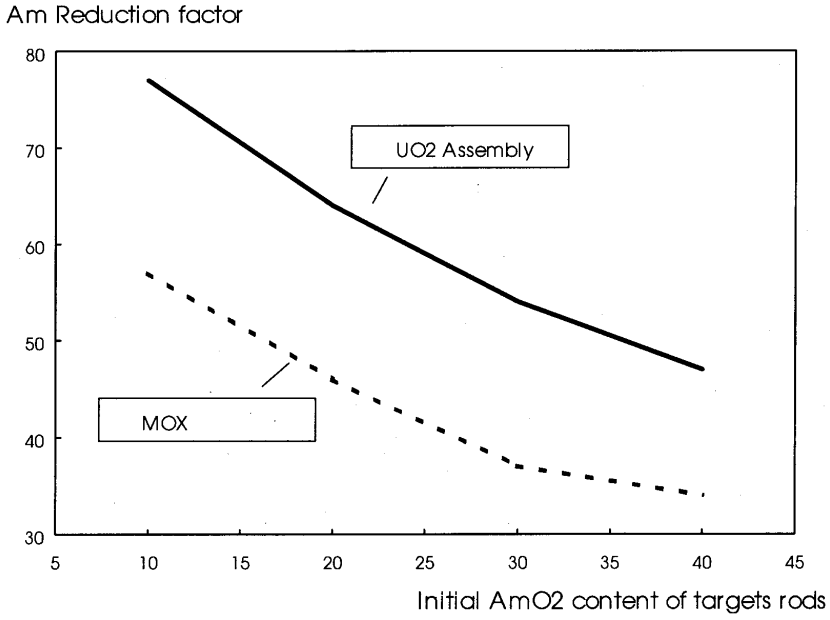
Heterogeneous recycling of Americium: detailed results

A practical heterogeneous concept

In the heterogeneous concept, Americium oxide is inserted in a matrix in the form of special target rods. The assumption is to mix AmO_2 and Al_2O_3 or UO_2; other support materials are currently envisaged, and Am_2O_3 could be envisaged as well, but as far as core physics is concerned, the present studies retain a sufficiently good orientation value.

Former core studies have shown that low fractions of 10 to 20% AmO_2 in volume would be optimal to transmute Americium (see Figure 2), but here this parameter is allowed to vary in order to accommodate the growing Am quantities.

Figure 2. **Heterogeneous recycling of Americium in PWR** (mass reduction after 44 GWD/t)



Reference [5] has shown the merits of two promising concepts, in which the Am target pins are placed, either in peripheral core assemblies, or in the corner pin positions of each MOX assembly. These two concepts are equally possible solutions with acceptable core performances and Pu+Am incineration properties. However, the MOX corner pin positions appear to fuel manufacturers and to core designers most practical. The neutron flux is also twice as high as in the peripheral assemblies. Detailed calculations have been performed for the Am targets in MOX corner positions, and their results are displayed below.

On Figure 1 with the sketch of the MOX assembly, the 12 corner pins, usually containing Pu with a lower enrichment, are now replaced by Am target pins. This is a good buffer zone between the UO_2 and MOX fuels so that the Am transmutation is relatively high, thanks to the thermalized spectrum of the UO_2 zone, see Figure 2. The Pu enrichments of the 2 remaining medium- and high-content MOX zones need to be re-adapted. It is assumed that the Am target pins stay in the core as long as the MOX pins, i.e. 4 years, before being reprocessed. When Pu and Am are recycled after reprocessing of spent fuel (including the Am targets), the raise in Am quantities is coped with a raising fraction of Am in the target pins for the successive MOX-X recycling steps.

But again, when using depleted Uranium support in MOX, the Pu enrichments would soon exceed the 12% enrichment limit. The depleted U is, therefore, replaced at first step by natural U, and further on by enriched U, so as to keep the Pu enrichment fixed at 8%. The same type of progressively enriched UO_2 support is used in the Am target pins.

Multiple recycling results

This (Pu+Am) multiple recycling strategy has been explicitly modelled by calculations, using a macrocell representing the MOX assembly with its UO_2 environment. After 15 recycles, the equilibrium is nearly reached for Pu and Am quantities : the unloaded amounts are equal to the loaded ones in the PWR system. The isotopic vectors, too, stay stable from any discharge to the next one.

Figure 3 illustrates the approach to equilibrium in the case where Pu alone is recycled, and Figure 4 when Pu and Am are simultaneously recycled. Tables I and II also show below, for the two cases, Pu only or Pu and Am, how are varying the fraction of MOX assemblies, the ^{235}U enrichment of the UO_2 support (it stays at 3.7% in the UO_2 assemblies), and the volume fraction of Am in the targets.

Figure 3. **Multiple recycling of Pu in MOX fuel with enriched ^{235}U support**
Increase of a) U enrichment b) MOX fraction in core loading

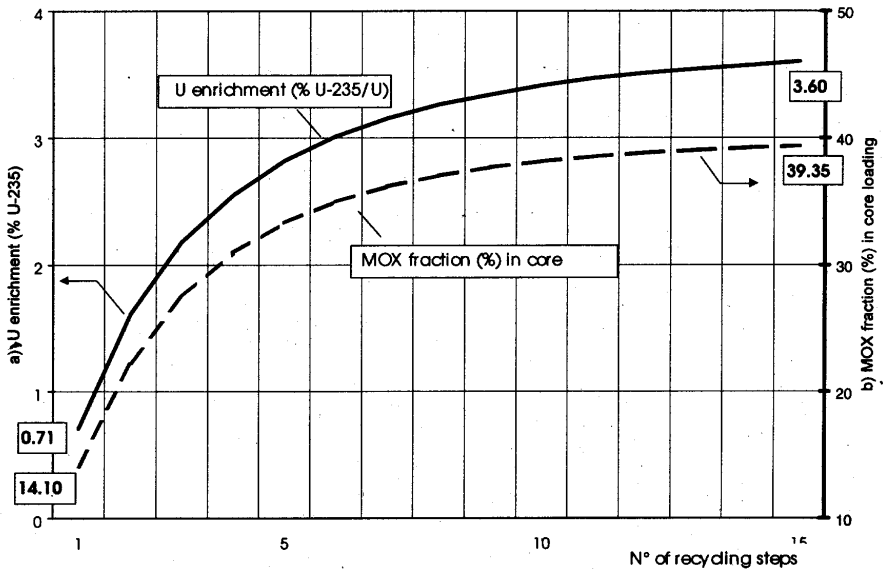


Figure 4. Multiple recycling of Pu and Am in targets with enriched ^{235}U support
Increase of a) U enrichment b) MOX fraction in core loading c) AmO_2 fraction in targets

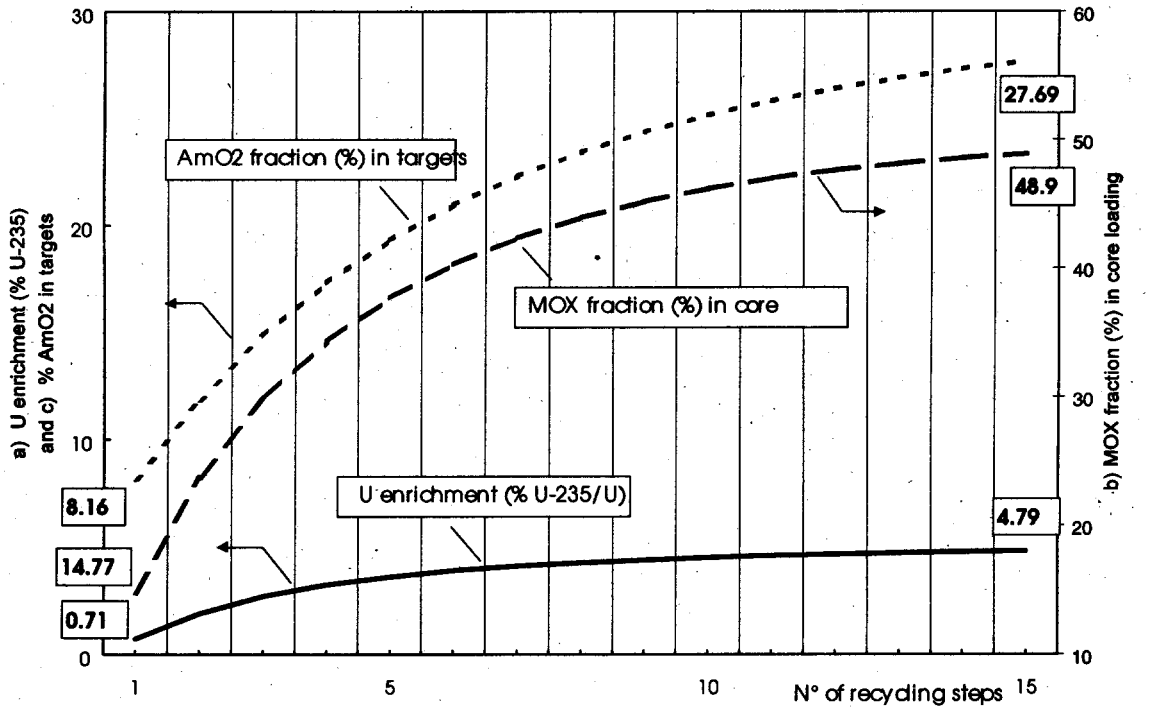


Table 1. **Multiple Pu recycling using enriched UO_2 in MOX**

Recycling steps	UO_2 enrichment in ^{235}U (%)	MOX fraction in core (%)
1	0.71	14.1
2	1.6	22.3
3	2.2	27.5
5	2.8	33.3
10	3.4	38.1
15 (equil.)	**3.6**	**39.4**

Table 2. **Pu+Am recycling using enriched UO_2 in MOX and target pins**

Recycling steps	UO_2 enrichment in ^{235}U (%)	MOX fraction in core (%)	Volume fraction of AmO_2 in targets (%)
1	0.71	14.8	8.2
2	1.9	23.7	11.9
3	2.7	29.8	14.9
5	3.6	37.7	19.3
10	4.5	46.2	25.1
15 (equil.)	**4.8**	**48.9**	**27.7**

The equilibrium between production and consumption of Plutonium and Americium needs:

- that about 50% of all assemblies be fuelled with MOX (vs 40% with Pu only).
- that the UO_2 support of the MOX assemblies be enriched to 4.8% in ^{235}U (versus 3.6% with Pu only).

The isotopic vectors at equilibrium are:

$^{238/239/240/241/242}Pu$ = 4.4/36.8/25.7/11.9/21.2 % (recycling Pu only),
or 11.1/34.9/23.3/10.9/19.8 % (recycling Pu+Am),
and $^{241/242/243}Am$ = 56.9/0.3/42.8 % (Pu only) or 43.6/0.5/55.9 (Pu+Am).

Table 3 gives for the UO_2 once-through cycle, and for the Pu and Pu+Am equilibrium cycle, the quantities of heavy material to be loaded or discharged per year at equilibrium for a yearly electricity production of 1 TWhe.

Table 3. **Mass balances for three recycling strategies**
(kg/yr corresponding to 1 TWhe)

Strategy	Once-through UO_2	MOX Recycling Pu only	MOX Recycling Pu+Am
^{235}U	460	429	480
Pu	130	363	432
Am	6.3	29.4	72.0
Cm	0.5	5.8	16.4

In Table III, the quantities in bold characters go to waste.

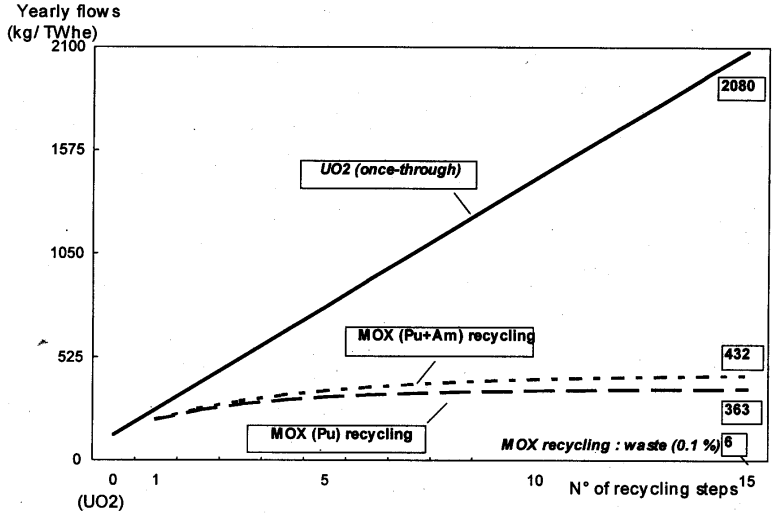
One can see the penalties incurred by recycling Am in addition to Pu: all mass flow rates have increased. In particular, the Cm masses are 3 times higher.

On the other hand, calculations have also checked that in these multiple recycling schemes, even when half of the assemblies are to be MOX ones, the power distributions can remain conform to the criteria throughout irradiation.

Waste radiotoxicity reductions

The success of these multiple recycling strategies in PWRs can easily be observed for what concerns the quantities of Pu and Am which are, either recycled and stabilised, or rejected as waste. As an example, Figure 5 shows that with the MOX (Pu) recycling strategy already, the success is significant with respect to the reference UO_2 once-through case. The MOX (Pu+Am) recycling strategy offers about the same type of success.

Figure 5. **Accumulation of Pu quantities rejected as waste**
rejected as waste as recycling proceeds

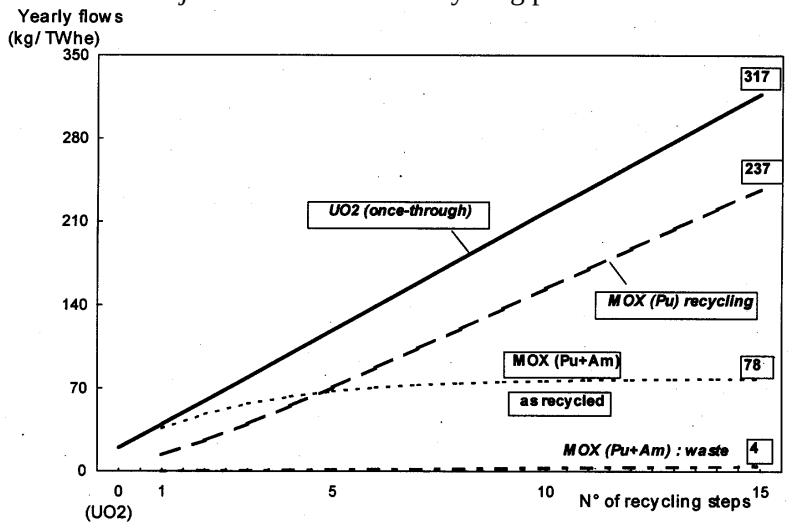


It is necessary to convert the quantities of the various Pu, Am and Cm isotopes into toxicities, to check what is effectively the reduction in terms of waste toxicities. One has of course to specify for what range of waste storage times one wants to reduce the toxicity.

Two typical storage times appear important: 1 000 and 10 000 years. For 1 000 years, the major sources of toxicity are known to come from the following isotopes: ^{241}Pu + ^{241}Am, then ^{240}Pu, ^{239}Pu and ^{243}Am. In the once-through case, these isotopes contribute as follows to the waste toxicity: ^{241}Pu+^{241}Am: 65%; ^{240}Pu: 20%; ^{239}Pu: 13%; ^{243}Am: 2%.

Figure 6 illustrates the relative effect of the MOX recycling strategies on the 1 000 year-toxicity by displaying the accumulation of the ^{241}Pu+^{241}Am quantities in the once-through case, and as recycling proceeds, in the two MOX (Pu, or Pu+Am) strategies. One can see that there is a remarkable success for the latter one, as could be easily predicted.

Figure 6. **Accumulation of ^{241}Pu + ^{241}Am quantities**
rejected as waste as recycling proceeds



The comparison after 10 000 years is less conclusive. Indeed, not only ^{239}Pu, ^{240}Pu and ^{243}Am are playing an important role in the toxicity, but also the raise in Cm quantities, as ^{244}Cm rapidly decays into ^{240}Pu and ^{245}Cm has a half-life of 8 500 years, comparable to that of ^{240}Pu (6 550 years).

Table 4 enables a judgement to be made on the compared quantities of ^{239}Pu+^{240}Pu+^{243}Am+^{244}Cm +^{245}Cm rejected as waste as an indicator of the waste toxicity after 10 000 years.

Table 4. **Indications on waste toxicity after a storage time of 10 000 years [quantities of $^{239+240}$Pu+^{243}Am+$^{244+245}$Cm (kg) rejected as waste after 15 recycling steps]**

UO_2 once-through	MOX (Pu) multiple recycling	MOX (Pu+Am) multiple recycling
1 701	168	209

For such a storage time, the MOXX (Pu+Am) recycling strategy is obviously less good than the simpler MOX (Pu) one. The reason for this paradox is given by the larger accumulation of Curium quantities.

In conclusion, the success of the MOXX (Pu+Am) recycling can be qualified, with respect to the MOX (Pu) one, as follows :

- Largely favourable for waste storage times around 1000 years.
- Not quite as good for waste storage times around 10000 years.

Fuel fabrication aspects

The homogeneous mixing of Am oxide into the usual (UO_2PuO_2) fuel was examined, with respect to the present fuel fabrication conditions at the BELGONUCLEAIRE MOX fuel plant [7]. Front-end operations, like the powder blending in the milling glove box, are most dose-intensive [8]. It was found that the combined recycling of Pu+Am corresponded to a substantial increase of the doses, by a factor 4.5. This can be coped with extra shielding layers, what complicates the fabrication and somewhat increases the cost of operations, but still seems a feasible extension of the standard MOX fabrication process.

In comparison, the fabrication of Am oxide target pins in a MOX fabrication plant in view either to feed the corners of MOX assemblies or to constitute dedicated assemblies, increases the doses by large factors, like typically 80 in front of the shielded doors of the oxide powder storage vaults, or even 2 800 from a canister transferring 200 pins to the mounting hall. This means that the fabrication of Am oxide targets should be done in shielded hot cells. More details can be found in [8].

Discussion and conclusions; further work

Summary of the main observations

Chemistry experts are presently requested to develop industrial ways to produce pure Am oxide from reprocessing. When this endeavour will materialise, the recycling of pure Am targets as considered above will be made possible, and the studies presented above will bear full significance.

From our orientation studies, both homogeneous and heterogeneous recycling schemes appear feasible. Multiple recycling of Pu+Am in PWRs is however possible only if enriched UO_2 is used as support of the MOX fuel. The fabrication of target pins should be hosted in hot cells rather than in glove boxes, so that fabrication costs are likely to be higher; the cost increase cannot be quantified yet.

The efficiency of stabilising total Am quantities in the whole cycle, is comparable in both cases. Target pins placed at MOX corners or in dedicated assemblies are both viable. The solution of MOX corners, presented in details here, is most effective, as the neutron flux is higher and the spectrum softer, what favours Am transmutation.

It is shown that recycling Am in addition to Pu leads to significant penalties: one third more ^{235}U is needed, the MOX fraction is increased from 40 to 50% in the PWRs, and there is a large raise of the Cm amounts. This problem of Cm deserves further studies.

While the multiple recycling of Pu+Am is effective to reduce the waste toxicity for storage times of about 1 000 years by a large factor (the MOXX (Pu+Am) strategy is 3 times better than the usual MOX strategy), there is no improvement but rather a slight deterioration for waste toxicities after 10 000 years. This is due to the enhanced production of Cm, by a factor 3, from Am irradiation.

Modified schemes for Am recycling

An alternate scheme which deserves to be investigated implies to keep the Am target pins 2 or 3 times longer in the reactor, so as to be entitled to reject these targets without further reprocessing. Obviously a certain part of the overcosts can be avoided in such a way. The balance is to be established, to check whether this prolonged irradiation can reveal globally beneficial. Parametric studies have already been launched, which indicate the potential interest of such a scheme, but full scale recycling calculations are still needed to confirm the advantages.

On the other hand, Am is not yet separated nowadays in an industrial scale from Cm and lanthanides at fuel reprocessing. These orientation studies have thus been complemented in order to know what would be the effect of accompanying Cm and lanthanides.

For a relatively small residual ratio of lanthanides on Am+Cm (e.g. 5 to 20%), these could be accepted as they do not penalise significantly core properties neither transmutation. The presence of Cm itself has a favourable influence on the power release of the targets, and therefore on their transmutation properties, which overcompensates the effect of lanthanides.

But the huge emission of alpha, gamma and neutron doses by Cm isotopes is known to be so severely hampering the handling of such powders that so far only gram quantities of Cm were treated, with the appropriate shielding. Hot cells are mandatory for any fabrication.

The fabrication of Am+Cm targets should thus be differed as long as realistically possible, e.g. by some 20 to 40 years, to let most of the Cm transform into Pu and ease this fabrication (anyway in automated cells of the fuel plant). The irradiation of these Am+Cm targets could then last longer than for the usual MOX pins, so as to be able to throw directly the spent targets to waste, avoiding any subsequent reprocessing.

,Explicit recycling calculations are needed to verify how feasible are in such schemes the handling of powders before and during target fabrication, and the behaviour of the targets under irradiation ; finally, the residual masses of Am and Cm transferred to the waste are to be assessed.

One advantage of the heterogeneous option is obvious: it makes possible to manage in two different ways the Pu in MOX pins, and the targets. For example, the recycling of Pu could be done with the shortest cooling times possible (3 + 2 years) so as to burn quickly the highly fissile ^{241}Pu, while the recycling of a mix of Am+Cm could be deferred to let Cm transform into Pu before irradiation. Also, the Am+Cm targets could be allowed to stay longer than the MOX pins, so as to avoid if possible further reprocessing.

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MINOR ACTINIDE DESTRUCTION IN DEDICATED REACTORS

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Abstract

The neutronic performance of small size critical dedicated burners is investigated here according to the type of recycling performed (MA supply alone or together with some fissile element), the nature of the possible fissile supply (Pu, enriched U, U bred from Th), the coolant type (sodium or lead).

The main neutronic parameters are given, such as reactivity coefficients, together with mass balances, MA consumption and fuel composition. The trends for future studies are also indicated.

Introduction

Several reactor concepts enabling to burn minor actinides (MA) have been considered. A fast neutron spectrum has been shown to allow a positive neutron gain throughout the burning process [1], making it theoretically independent from an external neutron supply (e.g. by fuel enrichment in fissile nuclides, or a spallation source). However, MA burning has important consequences on the core safety, fuel cycle and economic related parameters, besides neutronic feasibility only.

The fraction of electrical power supplied to the grid by the MA burners present in the reactor fleet is a key parameter for any MA burning scenario. This fraction can range from a theoretical value as low as ≈6% (only MA loaded in the burners, and heat to electricity conversion of 40%, as today fast reactors) to a full 100% (MA are homogeneously "sprinkled" over all the reactors in the fleet).

The design of specific MA burners, called *dedicated reactors* hereafter, with important MA loads (ideally 100% of the reactor fuel) would allow to restrict specific cycle operations (fabrication, handling, reprocessing) and safety countermeasures to a limited fraction of the reactor fleet with respect to the homogeneous option.

Main assumptions

The main lines followed by our dedicated reactor studies have been first an important reduction in core size from conventional fast reactor designs, in order to enhance leakage effects in case of coolant voiding, as the presence of MA has a strong adverse effect on this parameter. The typical sizes retained for this study are in the range from 100 to 300 MW_{th}. Secondly a dense and "cold" fuel (i.e. with an important thermal conductivity) has been retained: the nitride fuel. Finally, a multiple recycling has been modelled in order to account for possible important changes between the first and equilibrium recyclings.

Within this frame, several parameters were varied, such as the type of recycling (i.e. whether MA alone or mixed with some fissile material are added to the spent fuel from a recycling to the next), the nature of this possible fissile supply (i.e. Plutonium, enriched Uranium, Uranium bred from Thorium), the coolant nature (sodium or lead), the possible use of a moderator within the core ($^{11}B_4C$, $ZrH_{1.8}$).

Tools and methods

The code system ERANOS1.2 has been used throughout this study to assess the neutronic parameters of dedicated cores [2]. Basic nuclear data are taken from the ERALIB1 library, containing data from JEF2.2 evaluations adjusted on a wide set of experimental configurations (≈350, but covering oxide fast reactor cores, standard or CAPRA-type ones, *not* dedicated small reactors with large amounts of minor actinides, non-oxide fuel, moderator within the core...).

For the first scoping studies, the calculation route was rather a crude one: homogeneous cell calculations with a fine (1968) group energy mesh providing by condensation the 33-group cross-sections used in whole core calculations, which were performed in diffusion theory. This preliminary range of calculations was only meant to provide trends in order to select a fewer number of cases on which to perform more refined calculations.

This second range of calculations has been performed with heterogeneous cell models, either two-zone (fuel in the inner zone ; steel, coolant and moderator mixed in the outer zone) or a full subassembly (S/A) model, the individual pins and wrapper being then explicitly taken into account. Then the whole core calculations were performed using transport theory using an S_n method (P_1, S_8). Once again, the cell calculations include a step in fine groups, and the core calculations use a condensed 33-group energy mesh.

A decay chain ranging from A=231 (^{231}Pa) to A=248 (^{248}Cm) has been used. The core burn-up is performed from beginning of life (fresh fuel) conditions to end of life (fully burnt fuel) conditions, i.e. no detailed batch management scheme has been investigated.

Two models of multiple recycling were used : the so-called *fertile* and *mixed* recyclings. A fertile recycling means that only MA are added as a topping material to the spent fuel of cycle n to form the loading of cycle n+1. Whereas in the mixed recycling MA are added together with some amount of fissile material (Pu, ^{235}U, ^{233}U).

The fertile recycling procedure can be detailed as follows : the spent fuel from cycle n is left to cool down for 5 years, then the fission products are removed and all the actinides recovered, with a 0.1% loss for Th, U and Pu, and a 1% loss for MA, going to the waste stream. Enough MA are added to obtain a given actinide mass (the same as the initial actinide mass of cycle 1). This homogeneous mixture, after a 2-year ageing, is then put in the core S/A to form the loading of cycle n+1. It is worth noting that in a fertile recycling, for a given fuel density in the S/A, the core size at equilibrium conditions is fixed by reactivity considerations : the reactivity must be closed to zero at the “end of cycle” conditions, i.e. at roughly half the fuel residence time with the simplified burn-up assumptions specified above.

The treatment of the mixed recycling is slightly more complex. The relative amounts of MA and fissile nuclides to be added to the spent fuel of cycle n are determined by three conditions: firstly, the total addition must restore a given initial actinide mass (that of the beginning of life conditions of cycle 1). Secondly, the ratio of the fissile (Pu, ^{235}U or ^{233}U) contents in the inner and outer core zones must be kept constant (for power flattening). Thirdly, the beginning of life reactivity of cycle n+1 must be the same as for cycle 1. The iterative algorithm used provides a unique solution to these requirements. In a mixed recycling the core size is no more a restrictive condition, provided that the necessary fissile topping required remains positive and lesser than the complement to the initial mass.

The coolant void reactivity is computed by fully voiding the core (fissile) zones, and keeping the reflector and shield regions full of their coolant. The Doppler effect is calculated by putting the actinide temperature from T_1 = 1500 to T_2 = 450 K, and the resulting *Doppler constant* is given by: $K_D = [k(T_1) - k(T_2)] / \ln(T_1/T_2)$.

Results

Results of the preliminary scoping studies

Here, core calculations were performed in diffusion theory, with a 33-group energy mesh, using data output by homogeneous cell calculations. A small core was used, modelled in a RZ geometry. The core height and radius were 60 and 66 cm respectively, with a thermal output of 250 MW. With a S/A pitch of 12.4 cm, this corresponds to 96 fissile S/A, divided in two fuel enrichment zones. The coolant is sodium, allowing for a "classical" S/A design.

A first result was that the reactivity loss in cores containing large amounts of MA is quite low, even for such small cores, allowing for large fuel residence times. A 1 500 equivalent full power days (efpd) residence time is assumed throughout the study.

The other main findings were that the use of oxide fuel made it difficult to perform an indefinite multiple recycling, while a denser fuel like nitride allowed it easily. Similarly the presence of an inert matrix mixed to the fuel impairs the recycling capability. Even with such a small core size, the coolant void reactivity remains quite important with a sodium cooling (and furthermore the delayed neutron fraction is lesser than in a standard fast reactor core). The Doppler constant remains quite low (hard spectrum) unless a very efficient (i.e. hydrogenated) moderator is added to the fuel in the form of separate pins. Finally the fuel composition at equilibrium is far from those of standard fast reactors. These points are illustrated in the following tables. Note that reactivities are measured in *pcm*, with *1 pcm = 10^{-5} Δk/kk'*. These tables are related to a mixed recycling with Pu coming from a UOX PWR spent fuel as a fissile material, and a recycling of Np+Am+Cm as MA. The moderator used is $ZrH_{1.8}$ representing a 5% volume fraction of the S/A.

Table 1. **Main parameters, 250 MW_{th} dedicated core, simplified scheme**

	First cycle	16^{th} cycle (quasi-equilibrium)
Average Pu content (w%)	46.2	63.2
Pu supply (kg/TWhe) MA supply (kg/TWhe)	– –	55.0 49.3
Reactivity loss (pcm/efpd) Sodium void worth (pcm) Doppler constant (pcm)	-7.63 1564 / 1675 -152 / -242	-7.80 1611 / 1587 -460 / -548
Delayed neutron fraction (pcm) Prompt neutron lifetime (10^{-7} s)	203 / 190 2.32 / 3.37	211 / 223 3.05 / 4.23
Max. Linear rating (W/cm) Max. Burn-up (%)	399 31.6	386

Values separated by a slash (/) are relative to beginning and end of life conditions. This convention will also hold for Tables 3, 4 and 5. Burn-up has here a small effect on the sodium void worth value because of a competition between the reduction in MA content and fission product build-up between beginning and end of life.

The fuel composition at the beginning of the equilibrium cycle is 4.4% U + 6.5% Np + 63.2% Pu + 17.0% Am + 8.9% Cm. The isotopic composition of Plutonium is:

Table 2. **Plutonium isotopic composition by mass, equilibrium cycle**

^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
17.2%	17.1%	38.9%	6.9%	19.9%

The fertile recycling scheme (i.e. supply of MA only) has been investigated, as it would allow the maximum MA consumption by fission (≈105 kg/TWhe).

In this case, the size of the core is determined by the need to have a well-balanced reactivity at equilibrium conditions. One is faced then to a dilemma : with no moderator within the core, the coolant void reactivity is small, but the Doppler feedback is vanishing, and to put some moderator within the core leads to increase the core size and so to reach large coolant void values. This is illustrated in table 3, with lead cooling instead of sodium (volume fraction in the fuel : 20-25% fuel, 5-0% moderator, 15% steel and 55% lead).

Table 3. **Core parameters, fertile recycling, with or without moderator**

Moderator	None	$ZrH_{1.8}$
Core power (MW_{th})	95	180
Core height/radius (cm)	45 / 47	55 / 66
Average burn-up (%)	18.9	17.9
Max. Damage (dpa)	164	128
Reactivity loss (pcm/efpd)	-4.59	-1.78
Coolant void (pcm)	-294 / -969	1548 / 659
Doppler constant (pcm)	-15 / -22	-157 / -208
Delayed neutron fraction (pcm)	143 / 156	154 / 172
MA feed (kg/TWhe)	107.1	106.9

Results of the refined scoping studies (RZ model)

These studies involve a more refined calculation scheme, i.e. transport calculation with heterogeneous cell model, see §3. This series of calculations involved a mixed recycling, with either a sodium or lead coolant, and different natures of the fissile supply (Pu from UOX PWR spent fuel, ^{235}U, ^{233}U). Results are summarised in tables 4 and 5 below. The core has a height of 50 cm and a radius of 53 cm, for a thermal output of 125 MW, and so an electrical output of 50 MWe. Twenty-one recyclings are performed to reach the equilibrium conditions, which are given in the tables.

Table 4. **Refined calculations, core parameters, sodium cooling**

Fissile composition	Pu	^{235}U	½ ^{235}U + ½ Pu	¼ ^{235}U + ¾ Pu	^{233}U
MA composition	MA	MA	MA	MA	0.9 MA + 0.1 Th
Average burn-up (%)	18.7	18.7	18.7	18.8	18.9
Max. Damage (dpa)	114	103	107	110	110
Reactivity loss (pcm/efpd)	-7.48	-14.61	-9.42	-8.30	-9.32
Coolant void (pcm)	1610/1698	-258/135	1089/1327	1394/1553	836/1049
Doppler constant (pcm)	-220/-270	-58/-50	-186/-233	-208/-255	-254/-339
Del. neutron fraction (pcm)	195/204	513/464	284/271	232/231	217/220
Prompt n. lifetime (10^{-7} s)	2.79/3.76	3.97/5.75	3.12/4.19	2.92/3.91	3.20/4.46
Fissile feed (kg/TWhe)	48.4	94.3	64.2	55.3	35.0
MA feed (kg/TWhe)	56.7	9.6	40.5	50.0	63.7

Sodium void values remain large with Pu as a fissile supply. Using enriched U (90% ^{235}U) instead of Pu leads to reduce the sodium void worth and to increase the delayed neutron fraction, but to the cost of a reduced Doppler feedback, of an increased reactivity loss, and of a quasi total loss of the MA burning capability (the U content in the core becomes very large).

Fissile supplies made of a mixture of U and Pu have thus been investigated, and allow to mitigate the drawbacks and advantages of pure U as fissile supply. However, the use of enriched U to operate dedicated cores in a long-term perspective can be questioned.

U bred from Th as fissile supply is also more attractive than Pu, but would require to start a Th cycle in conventional power reactors.

Table 5. **Refined calculations, core parameters, lead cooling**

Fissile composition	Pu	Pu	^{235}U	½ ^{235}U + ½ Pu	^{233}U
MA composition	MA	Am+Cm	MA	MA	0.9 MA + 0.1 Th
Average burn-up (%)	18.5	18.4	18.4	18.5	18.6
Max. Damage (dpa)	113	117	112	112	111
Reactivity loss (pcm/efpd)	-4.85	-4.71	-8.67	-5.83	-7.10
Coolant void (pcm)	1244/647	1485/829	-401/-356	869/421	70/-258
Doppler constant (pcm)	-203/-254	-328/-399	-134/-185	-188/-237	-273/-352
Del. neutron fraction (pcm)	179/191	163/182	318/291	222/219	204/210
Prompt n. lifetime (10^{-7} s)	2.76/3.76	3.01/4.11	3.31/4.56	2.93/3.95	3.40/4.67
Fissile feed (kg/TWhe)	26.9	21.9	49.3	34.1	24.0
MA feed (kg/TWhe)	78.8	83.8	55.7	71.4	73.4

It can be seen in table 5 that lead cooling allows reduced coolant void values and larger MA consumptions than sodium cooling, and that the use of ^{235}U as fissile supply has a lower negative impact on Doppler feedback and MA consumption.

Table 6 on next page gives the fuel isotopic compositions for the two cases in table 5 involving Plutonium as a fissile supply. Here the slash (/) separates compositions at the beginning of the first and equilibrium cycles. The complement to 100% is Uranium. This table shows the large Pu content in the fuel at equilibrium (≈55%) and its much degraded isotopic composition.

Always for a Pu fissile supply, the proportion of power coming from dedicated MA burners in the global reactor fleet power output ranges from 6% (Am+Cm burners) to 8% (Np+Am+Cm burners). This is quite low, but one must keep in mind the large number of small power individual burner reactors required to reach this proportion, that is roughly 2 dedicated 50 MWe burner every 1 large-size (1 500 MWe) power reactors. This motivates investigations for larger dedicated core sizes, even if several small dedicated cores can be grouped as modules in a same plant.

Table 6. **Refined calculations, lead cooling: fuel compositions (%)**

MA feed	Np+Am+Cm	Am+Cm
^{238}Pu	12.65 / 16.14	8.68 / 11.03
^{239}Pu	8.08 / 6.93	6.04 / 4.97
^{240}Pu	20.63 / 21.90	23.80 / 24.99
^{241}Pu	2.50 / 3.29	2.62 / 3.67
^{242}Pu	6.89 / 7.94	7.77 / 9.11
^{237}Np	10.77 / 7.81	0.56 / 0.57
^{241}Am	11.70 / 8.32	16.27 / 11.04
^{242m}Am	0.66 / 0.80	0.90 / 1.08
^{243}Am	9.94 / 8.00	14.27 / 11.00
^{243}Cm	0.09 / 0.11	0.14 / 0.16
^{244}Cm	7.26 / 9.11	10.90 / 13.53
^{245}Cm	1.85 / 1.98	2.69 / 2.85
^{246}Cm	0.83 / 1.03	1.25 / 1.56
^{247}Cm	0.12 / 0.14	0.18 / 0.22
^{248}Cm	0.04 / 0.05	0.06 / 0.07

Comparison with more detailed calculations

Table 7 compares the results for the first recycling between two different cell models: a 2-zone cylindrical model and a full representation of the S/A, with individual fuel and moderator pins being represented. In both cases, the core model is cylindrical (RZ), and flux calculations are performed in transport theory (S_n method). The fissile is a mixture of 50% Pu + 50% ^{235}U.

Table 7. **Comparison of calculational models**

Cell calculation	2-zone	Full S/A
Fissile content (%)	49.8	48.1
Reactivity loss (pcm/efpd)	-8.11	-7.92
Sodium void worth (pcm)	1 476/1 780	1 383/1 664
Doppler constant (pcm)	-113/-147	-107/-137
Delayed neutron fraction (pcm)	207/192	200/187
Prompt neutron lifetime (10^{-7}s)	2.42/3.28	2.35/3.16

As can be seen, the differences are small, less than 8% on all parameters, and allow to use a 2-zone cell model, involving much less calculation time than a full S/A representation.

Trends for future work

Future studies will cover several topics. Some investigations will be performed toward a size increase for the MA burners. A 3D (Hex,Z) core modelling, using nodal transport methods will be used to assess more accurately the retained designs. Transient studies will be carried out to assess the behaviour of such cores during incidental sequences. Finally, an uncertainty assessment will be necessary as MA are among the main contributors to reactivity in dedicated, with not so well known nuclear data for them. The uncertainty calculations will cover the fields of core reactivity, reactivity loss , actinide inventory during burn-up.

Conclusions

The dedicated core optimisation process involves contradictory trends. To increase the MA mass consumption rates and to enable an indefinite recycling, one has to turn to dense fuels, to replace sodium cooling by lead cooling, and to perform a fertile recycling (MA supply only). On the other hand, the use of an efficient (hydrogenated) moderator is necessary to recover a significant Doppler feedback, and small core sizes favour a low coolant void : these points favour a mixed recycling (MA + fissile material supply).

The scoping studies performed will allow to carry out transient studies, in order to check the kinetic behaviour during incidental sequences, and an uncertainty analysis, to assess the impact of the poor knowledge of MA cross-sections on reactivity, burn-up, reactivity coefficients.

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STATUS OF TRANSMUTATION STUDIES IN A FAST REACTOR AT JNC

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Abstract

This paper presents the status of studies on transmutation of minor actinides (MA) and long-lived fission products (LLFP) in a fast reactor at JNC, which includes four areas of work: (a) design studies, (b) nuclear data measurement and evaluation, (c) fuel fabrication and irradiation test, (d) strategy studies. It was found that the mixed hybrid MA-loading method, where Np nuclide is dispersed uniformly in the core and target subassemblies containing Am, Cm and rare earth nuclides are loaded into the radial blanket region, has a great potential to achieve effective transmutation of MA without serious drawbacks in terms of core performance. Fission cross section ratios of minor actinide nuclides (^{237}Np, ^{241}Am, ^{243}Am, ^{242m}Am, etc.) in the fast and epithermal neutron energy region have been measured to evaluate the accuracy of MA nuclear data. As for fabrication and investigation of irradiation behaviour of MOX containing MA, the systematic program has been planned. The irradiation test of Np- and Am-contained MOX fuel is planned in JOYO. In step with the JOYO MK-III schedule, the irradiation test will be initiated from around 2003.

Introduction

Japan Nuclear Cycle Development Institute (JNC) was organised to establish the nuclear fuel cycle in Japan in 1st October 1998. JNC is now conducting research and development on FBRs and nuclear fuel reprocessing technology to achieve this objective.

One of the distinctive features of a fast reactor is its good neutron economy. Utilising the excess of neutrons enables us to construct flexible cores such that they incinerate minor actinides(MAs) and long lived fission products (LLFP) to reduce radiotoxicity and breed or burn Plutonium in consideration of Plutonium balance.

Some of the MA nuclides (Np, Am, Cm) contained in residual waste from reprocessing have extremely long-lived radiotoxicity [1]. Means of reducing the radiotoxicity of the MA nuclides are presently under investigation. The MA nuclides could produce useful energy if converted into short-lived fission products by neutron bombardment. From this standpoint, a nuclear reactor provides the obvious means for transmutation of MA nuclides. Among the various nuclear reactors, a fast reactor is considered to have the greatest potential to transmute MA effectively, because of its hard neutron spectrum [2-6].

Transmutation of ^{99}Tc and ^{129}I by neutron capture as a result of irradiation in nuclear reactors will yield the stable isotopes ^{100}Ru and ^{130}Xe, respectively. However, due to the small neutron cross sections, the transmutation efficiency in LWRs is low. Moderated subassemblies in fast reactors are more appropriate devices for the transmutation of the fission products [6-8].

This paper presents the status of studies on transmutation of minor actinides (MA) and long-lived fission products (LLFP) in a fast reactor at JNC , which includes four areas of work : (a) design studies, (b) nuclear data measurement and evaluation, (c) fuel fabrication and irradiation test, (d) strategy studies.

Feasibility studies on MA transmutation

Feasibility studies have been performed to investigate the basic characteristics (transmutation rate, burn-up reactivity, Doppler coefficient, sodium void reactivity, maximum linear heat rate, etc.) of a fast reactor core with MA transmutation, the following items were considered:

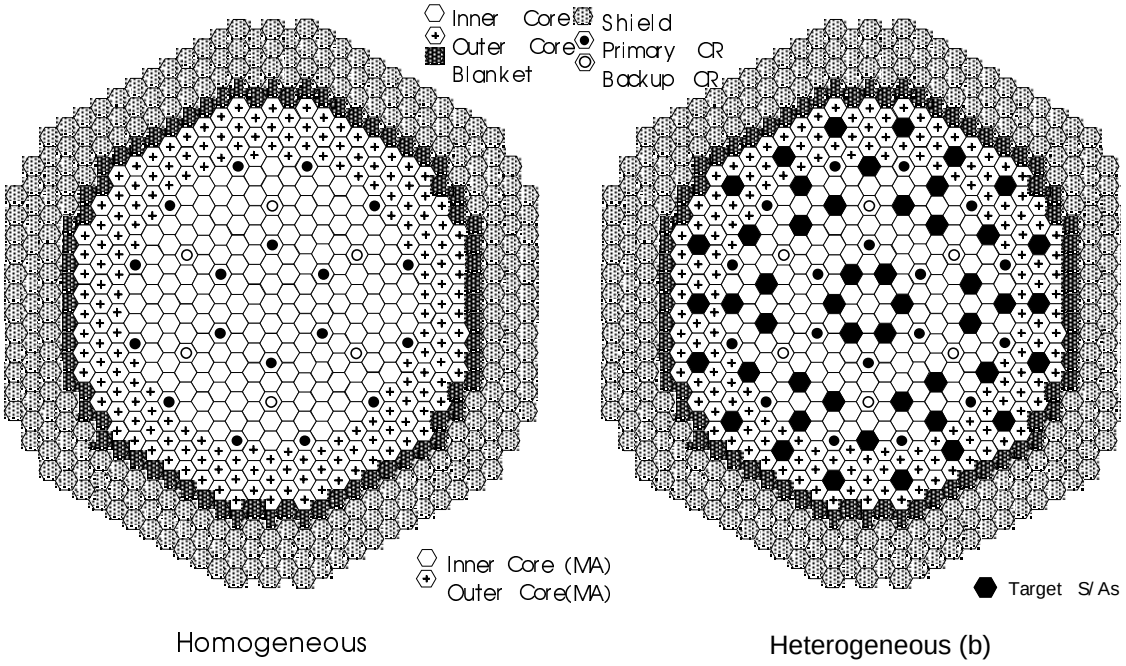
1. Study on loading method of MA in the core (homogeneous, heterogeneous, hybrid, blanket, etc.).
2. Selection of fuel material for MA transmutation (oxide, inert matrices such as Al_2O_3, CeO_2, etc.)
3. Study on the maximum tolerable amount of rare earth (RE) nuclides.
4. Effect of MA recycling on core characteristics and fuel cycle system.
5. Influence of uncertainties of MA nuclear data.
6. Influence of MA containing fuel on reactor plant and fuel cycle.

The main results of the studies are summarised as follows.

Study on MA loading method

The MA transmutation in a fast reactor core has no serious drawbacks in terms of core performance, provided that the homogeneous loading methods shown in Figure 1 can be employed with a small ratio of MA to fuel (~5% wt). Since a 1 000 MWe-class LWR produces about 26 kg of MA per year, a fast reactor with 5%wt MA loading can transmute the MA mass from six LWRs.

Figure 1. **MA loading method**



The heterogeneous MA loading method as shown in Figure 1 can be made feasible by optimising the fuel design, loading pattern and the coolant flow of the MA-loaded fuel subassemblies. The reduction of the fuel pin diameter and the Pu enrichment is essential to reduce the power of MA-loaded fuel in the heterogeneous MA loading method.

The hybrid MA loading method, where Np nuclide is dispersed uniformly in the core and target subassemblies containing Am, Cm and rare earth nuclides are loaded into the radial blanket region, can transmute a large amount of MA without serious drawbacks in terms of core performance. The transmuted mass of MA is about 530 kg/cycle as shown in Table 1, which is almost 16 times the mass produced by an LWR of the same power output.

The MA loading in the blanket region causes no problems from the viewpoint of core performance. Minor actinides are transmuted at a rate of 6% per cycle in the axial and radial blanket regions.

It was found that the hybrid MA loading method has the potential to achieve the maximum transmutation of MA with no special design considerations.

Table 1. **Comparison of core performance for various MA loading methods**

Item	Reference (No MA)	Homogeneous loading	Heterogeneous loading	Homogeneous loading	Hybrid loading
MA and RE loaded in the core region	–	Np,Am,Cm: 5% RE: 0%	Np,Am,Cm: 49% RE: 0% (Number of target S/As:39)	Np,Am,Cm: 5% RE: 10%	Np: 9.8% RE: 0%
Matrix of target	–	–	UO_2	–	Al_2O_3
MA and RE loaded in the blanket region	–	–	–	–	Am,Cm: 46% RE: 46% (Number of target S/As: 72)
Core height (cm)	100	100	100	100	100
Cycle length (days)	456	456	456	456	456
Number of batches	3	3	3	3	3
Pu enrichment (inner core/ outer core) (wt%)	15.4/18.6	16.6/20.1	15.4/18.6	20.0/24.2	19.0/23.4
B.U. reactivity loss (%δk/kk')	3.31	2.12	1.83	3.71	0.90
Max. linear heat rate (driver/target) (W/cm)	420	407	439/309	413	406/174
Void reactivity	1.0	1.3(1)	1.3(1)	1.4(1)	1.5(1)
Doppler coef.	1.0	0.6(1)	0.7(1)	0.5(1)	0.45(1)
MA transmutation Amount (kg/cycle)	–	172	186	164	529
Rate(%/cycle)	–	10.9	11.3	10.3	

(1) Relative values.

Selection of fuel material for MA transmutation

Different types of inert matrices, instead of UO_2, for the heterogeneous MA-loading method have been investigated, they avoid the buildup of higher actinides via ^{238}U and achieve a high MA transmutation rate. Inert matrices of Al_2O_3 and CeO_2 were examined in this study. The MA transmutation rate of the target subassembly using inert matrices is larger than that of the target subassembly using UO_2 The use of inert matrices in the target subassembly effectively increases the MA transmutation rate.

Study on the permissible re level in homogeneously loaded MA

Systematic parameter survey calculations were performed to investigate the basic characteristics of a fast reactor core loaded homogeneously with MA which contains RE, and also to establish a MA and RE loading method which has no serious influence on the core design. The homogeneous loading of MA and RE has no serious effects on the reactor core performance, provided that the amounts of MA and RE in the fuel are less than 5 and 10wt% respectively. In the case of adding Am, Cm and RE in the radial blanket region, it is possible, from the viewpoint of core performance, to insert ~50wt% of Am and Cm, and ~50wt% of RE in the target assemblies.

Effect of MA recycling on core characteristics and fuel cycle system

The effects of MA recycling on the core characteristics and the fuel cycle system in the homogeneous loading method were evaluated. The recycling of MA in a fast reactor is feasible from neutronic and thermal-hydraulic points of view. However, during multiple recycling the Np fraction is significantly reduced compared to the unirradiated feed, and the fraction of Cm is greatly increased because of neutron capture in Am. The accumulation of Cm as a result of the MA recycling will bring about some problems concerning fuel handling and reprocessing, because of an increase in both the decay heat and the neutron emission rate from ^{244}Cm.

Influence of uncertainties of MA nuclear data

The influences of the uncertainties upon nuclear characteristics were evaluated for a large LMFBR core loaded with MA of 5%. Sensitivity analysis on cross sections was carried out and uncertainties of nuclear characteristics were roughly evaluated. Uncertainties of nuclear characteristics are rather large compared with those of conventional cores. Some cross sections of minor actinides (^{237}Np, ^{241}Am, ^{238}Pu, ^{243}Am and ^{244}Cm) need to be improved.

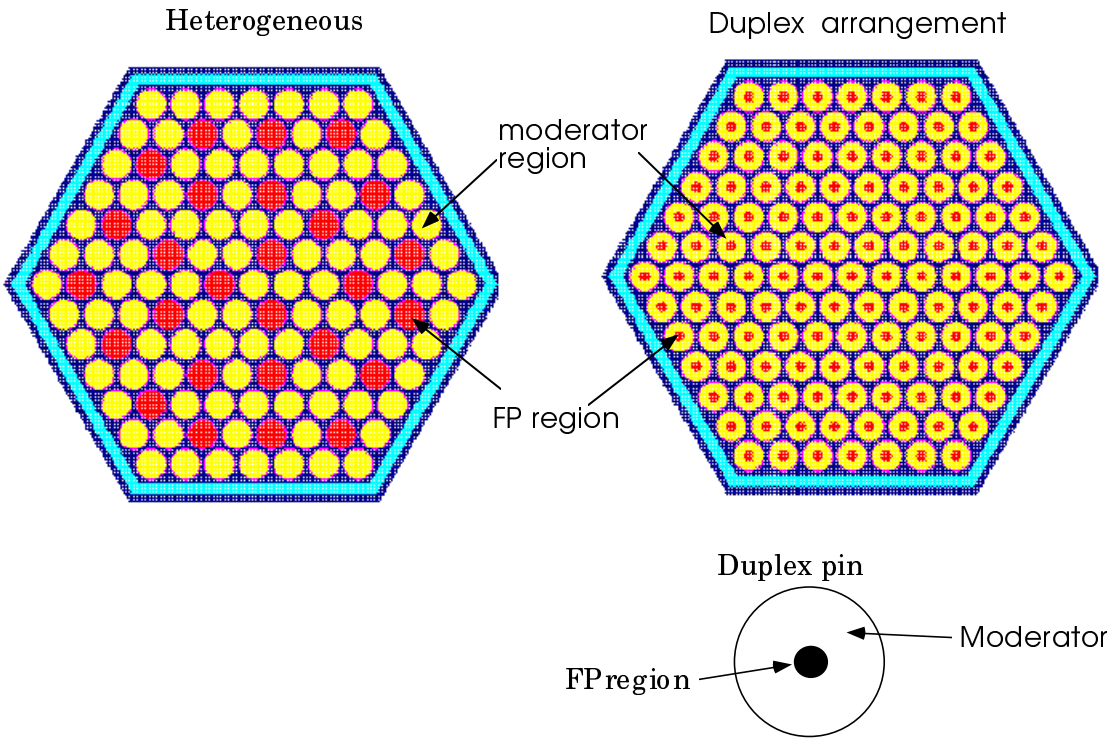
Influence of MA containing fuel on reactor plant and fuel cycle

Both the decay heat and neutron emission rate of the MA (Np, Am and Cm)-loaded fuel are very large in comparison with MOX fuel without MA. The dominant element of these fuel properties is Cm. If it is possible to remove Cm from the MA-loaded fuel, the decay heat value will decrease by one order, and the neutron emission rate by three orders. Since the dominant isotope, ^{244}Cm, has a relatively short half-life of 18 years, there might be another possibility of the fuel cycle, that is, partitioning of Cm and Am from MA in the reprocess and storing of Cm and Am for a period. Some study will be needed to estimate the trade-off between the plant modification and the reprocessing.

Feasibility studies on FP transmutation

A moderated target subassembly was used for FP transmutation. The subassembly consists of moderator pins and FP target pins distributed between the moderator pins. The moderated target subassemblies were loaded in the radial shield region of the fast reactor. A new concept of duplex pellets was also examined: a moderator annulus surrounding a ^{99}Tc core, as shown in Figure 2, adopted to get a better ^{99}Tc transmutation performance.

Figure 2. **Configuration of moderated target sub-assemblies**



Systematic parameter survey calculations were performed to investigate the basic characteristics of FP transmutation in the blanket region of a fast reactor. The arrangement of the moderator and the target pins in the subassembly, the moderator material and the volume ratio of target to moderator were selected as parameters. The results of the calculations are shown in Table 2. The transmutation rate of ^{99}Tc in the new target subassembly is higher than that in the subassembly consisting of separate $ZrH_{1.7}$ moderator pins and ^{99}Tc target pins, as shown in Figure 2. A maximum ^{99}Tc transmutation rate of about 10%/year was obtained by using the new target subassembly loaded in the blanket region of the fast reactor. The new target subassembly can achieve an optimum transmutation performance by adjusting the volume ratio of $ZrH_{1.7}$ to ^{99}Tc in the duplex pellet.

Table 2. **Results of ^{99}Tc transmutation performance parameter survey**

Loading method of FP pins	Number of pins in sub-assembly	Number of FP pins	Radius of FP pin	Transmuted amount (kg/y)	Transmutation ratio (%)
Heterogeneous	127	37	0.5	41.1	1.8
Heterogeneous	127	22	0.5	27.2	2.5
Duplex	127	127	0.2	38.1	3.5
Duplex	127	127	0.063	10.8	9.8
Duplex	217	217	0.2	46.7	2.5
Duplex	217	217	0.063	17.1	9.1

The effects on main core characteristics of loading target subassemblies were also analysed. It was found that the power density of the core fuel adjacent to the target is rather high and is about the same as the maximum in the core. However, the power spike is much mitigated compared to the case of loading target subassemblies in the core region.

Several calculations were performed to determine the ^{129}I transmutation performance. ^{129}I was loaded as NaI. The isotopic concentration of ^{129}I was 76.5% and the remainder ^{127}I. The transmutation rate of ^{129}I was 5.2% and the transmuted amount was 18 kg in a year. The amount of ^{129}I produced by a 1 000 MWe class PWR is about 5 kg, so the transmuted amount of ^{129}I was equal to the output from 3 PWRs.

Measurements of nuclear data for MA and FP transmutation

In MA burner core analyses, nuclear data for MA nuclides and fission products are of primary importance. However, nuclear data for many MA nuclides are still not known to the desired accuracy. Accurate experimental data of neutron cross section for MA are indispensable to establish MA transmutation technology by FBRs.

Fission cross section of MA nuclides

Fission cross section ratios of minor actinide nuclides (^{237}Np, ^{241}Am and ^{243}Am) relative to ^{235}U in the fast neutron energy region have been measured to evaluate the accuracy of MA nuclear data, using a back-to-back (BTB) fission chamber at YAYOI fast neutron source reactor. The experimental results were compared with the fission cross sections in the JENDL-3.2, ENDF/B-VI and JEF-2.2 libraries. It was found that calculated values for ^{241}Am using the JENDL-3.2, ENDF/B-VI and JEF-2.2 data are higher by 19%, 21% and 18%, respectively, than the measured value in the centre of the core.

Making use of BTB fission chambers and a lead slowing-down spectrometer coupled to a 46MeV electron linear accelerator at Kyoto university, the fission cross sections of ^{237}Np, ^{241}Am, ^{242m}Am, ^{243}Am have been measured relative to that for ^{235}U (n,f) reaction in the energy range from 0.1eV to 10 keV. Each of the fission cross sections in the JENDL-3.2 and ENDF/B-VI libraries was compared with the measurement.

Capture cross section of MA nuclides

The keV-neutron capture cross sections of ^{237}Np were measured to evaluate the accuracy of the nuclear data libraries using the 3-MV Pelletron accelerator of the Research Laboratory for Nuclear Reactors at the Tokyo Institute of Technology (TIT). The measurement was relative to the standard capture cross sections of ^{197}Au. A neutron time of flight method was adopted with ns-pulsed neutron source the accelerator and a large anti-Compton NaI(Tl) gamma-ray detector.

As a results, the capture cross sections of ^{237}Np were obtained with the error of about 4% in an incident neutron energy region of 10 to 500 keV. The present data are compared with other experimental data and the evaluated values of JENDL-3.2, and it was found that JENDL-3.2 provided good evaluations for ^{237}Np.

Decay heat of MA nuclides

The decay heat power of nuclear reactors is an important quantity for design, safety and costs of operation of a nuclear reactor. Extensive works have been done for decay heat measurements of major nuclides such as ^{235}U, ^{238}U, ^{239}Pu and ^{241}Pu and the accuracy has been established. No experimental decay heat data exist up to now for minor actinides such as ^{237}Np, ^{241}Am and ^{243}Am that are important for MA burner design.

The objective of this study is to measure the decay heat of minor actinide nuclides irradiated by the fast neutron energy spectra accurately in order to verify the decay heat calculation and thus establish confidence in the calculational method.

The sample irradiations of ^{235}U (reference) and ^{237}Np were performed in the fast neutron source reactor YAYOI of Tokyo University. Gamma and beta decay heat released from fission products of the samples were measured using the radiation spectrometry method. A NaI(Tl) scintillation detector was used for measurement of gamma ray. A plastic scintillation detector and a proportional detector were used for beta ray measurement.

Capture cross section of RE nuclides

While it is essential to consider all actinide containing wastes in a total actinide recycling scheme, the high-level waste (HLW) certainly presents the most difficult partitioning problem. Especially, separation of the trivalent minor actinides (MA) (Am, Cm, and higher products) from rare earth (RE) nuclides is notoriously difficult. Several methods have been successfully applied to isolating an actinides/RE fraction from the HLW. The impact of RE nuclides in MA containing fuels on the core performance of FBRs was investigated. As a result, it was found that the effect of RE nuclides on core characteristics is large. Accurate neutron cross section data of RE nuclides become necessary for designing the MA burner core. The data, however, are quite inadequate both in quality and in quantity.

Measurements of keV-neutron capture cross sections of RE nuclides (^{147}Sm, ^{148}Sm, ^{150}Sm, ^{140}Ce, ^{141}Pr, ^{153}Eu, ^{143}Nd, ^{145}Nd) have been performed to evaluate the accuracy of the nuclear data libraries using the 3-MV Pelletron accelerator of TIT. The capture cross sections of RE nuclides were obtained with the error less than 5% in an incident neutron energy of 10 to 500 keV.

A comparison was made between the present experimental data and the evaluated values of JENDL-3.2. The comparison shows that the JENDL-3.2 overestimates the keV-neutron capture cross sections of ^{147}Sm and ^{150}Sm by 5 to 15% and 5 to 20%, respectively, underestimates those of ^{140}Ce, ^{141}Pr and ^{153}Eu by about 30%, 5 to 20% and about 10%, respectively, and estimates those of ^{148}Sm very well.

Capture cross section of FP

The thermal neutron capture cross section and the resonance integral of radioactive fission products have been measured. For the measurement of thermal neutron cross section, new method named isotope ratio method has been developed. The thermal neutron capture cross section and resonance integral has been measured for ^{99}Tc, ^{129}I and ^{135}Cs [9-11].

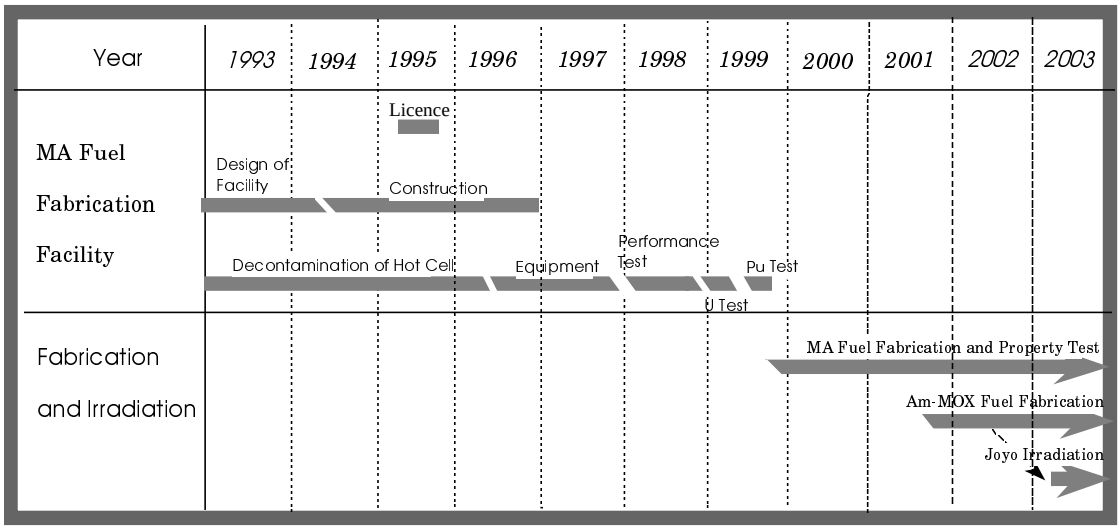
Transmutation rate

As a part of MA nuclear data evaluation, the analysis of irradiated ^{237}Np sample in Joyo has been performed. Additional irradiation test of Np, Am and Cm samples in JOYO was started in August, 1994.

Fabrication of MOX fuel containing MA

As for fabrication and investigation of irradiation behaviour of MOX containing MA, the systematic program has been planned in JNC. Two fabrication methods, pellet and vibro-packing have been studied for Neptunium-based fuel pins. The pellet type Np-based fuel will be fabricated at Tokai Works of JNC, and the fabrication of Np-based fuel by vibro-packing method will be performed at PSI in collaboration with JNC. For Am-based fuels, the Alpha-Gamma Facility (AGF) at Oarai Engineering Centre of JNC has already been reformed to fabricate MOX fuel pins containing Am at first as shown in Figure 3 and then containing Am and Np. Remote assembling will be conducted in the Fuel Monitoring Facility (FMC). Both facilities will provide test beds for the post irradiation examination. The irradiation test of Np- and Am-contained MOX fuel is planned in JOYO. In step with the JOYO MK-III schedule, the irradiation test will be initiated from around 2003.

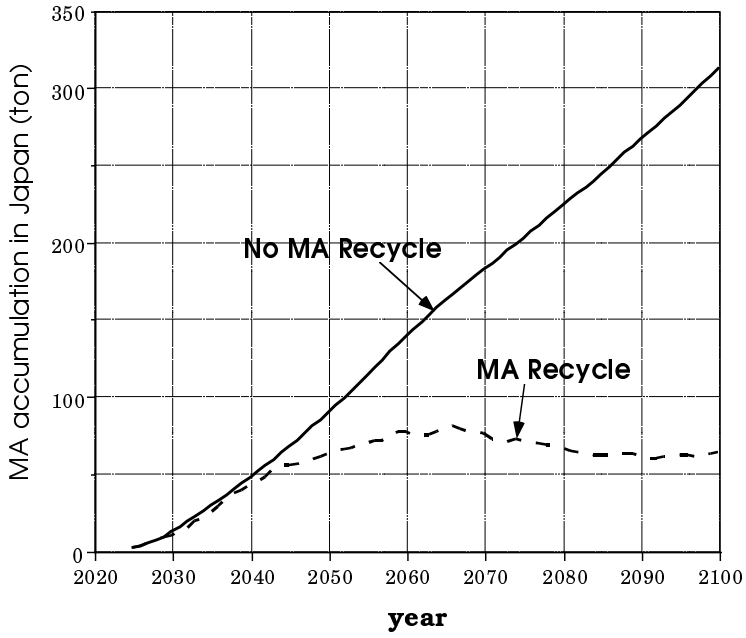
Figure 3. **Fabrication of MOX fuel containing Am**



Mass balance of MA in Japan

The MA mass balance was analysed according to the predicted nuclear energy production in Japan. Plutonium and MAs are recovered from the LWR and Pu-thermal reactors, recovered Pu and MAs are multiply recycled in fast reactors. Nuclear power generation is assumed to increase to 1 000 MWe/y, with the introduction of commercial fast reactors starting in the year 2030. New reactors are assumed to be totally FBR, and all spent fuel discharged from LWR and Pu-thermal reactors is assumed to be reprocessed. The total MAs transferred into the high level waste are calculated to be 310 tons from LWR, Pu-thermal LWR and FBR without recycling. In the case of recycling MAs into LMFRs after the year 2030, the MAs remaining in the fuel cycle in the year 2100 is reduced to about 60 tons, 80% less than without recycling , as shown in Figure 4 below.

Figure 4. **Effect of transmutation in reducing accumulation MA**



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MASS BALANCE OF THE PU AND MINOR ACTINIDES RECYCLING METAL FUEL SYSTEM

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Abstract

The mass flow of Plutonium and minor actinides is calculated for a future LWR-FBR fuel cycle model, in which a certain scale of power generation by LWRs is continued for a long period before the replacement by FBRs begins. The burnup of the LWR spent fuel is considered to be higher than the current standard. It is assumed that all the Plutonium and minor actinides recovered from LWRs are kept and used to start-up and feed metal fuel commercial FBRs. The results show that the accumulated Plutonium and minor actinides from the LWRs can be consistently consumed without further accumulation, by gradually establishing the same scale of the FBR power generation and its fuel cycle. The optimum content of the minor actinides in the standard FBR fuel is about 2 weight percents. If the FBR era is to come in the future, the extended LWR era causes no significant problem in terms of the consumption of the accumulated transuranic.

Introduction

Recovering minor actinides from the LWR spent fuel and utilising them in FBRs is expected to reduce the long-term radiotoxicity in the nuclear waste. It is proposed to recycle the minor actinides from the LWRs into metal fuel FBRs and their fuel cycle together with the recovered Plutonium [1]. Metal fuel FBRs show suitable performances for the consumption of minor actinides, and their pyro-reprocessing is quite compatible with the recycle of the minor actinides. In this case, the minor actinides content in the FBR feed is limited to as low as a few weight percents to keep the core safety parameters acceptable. A low content is also preferred to make a simple casting fuel fabrication method feasible [2]. But it is shown that the consumption of all the transuranic elements, i.e. Plutonium and minor actinides, from the LWRs is possible by introducing them widely into the commercial FBR fuel cycle.

However, the introduction of FBRs is being delayed recently. It causes the accumulation of the transuranic from LWRs. The relative amount of the minor actinides becomes larger during the extended decay period. Pursuing higher burnup of the LWR fuel can be another source for increasing the pressure of minor actinides accumulation.

In this study, considering these circumstances, the feasibility of the concept of the transuranic consumption by the commercial FBR fuel cycle is examined by evaluating the mass flow of the Plutonium and minor actinides for an LWR-FBR fuel cycle model. In this model, a certain scale of power generation by LWRs is continued for a long period before the replacement by FBRs and the consumption of the accumulated transuranic begin.

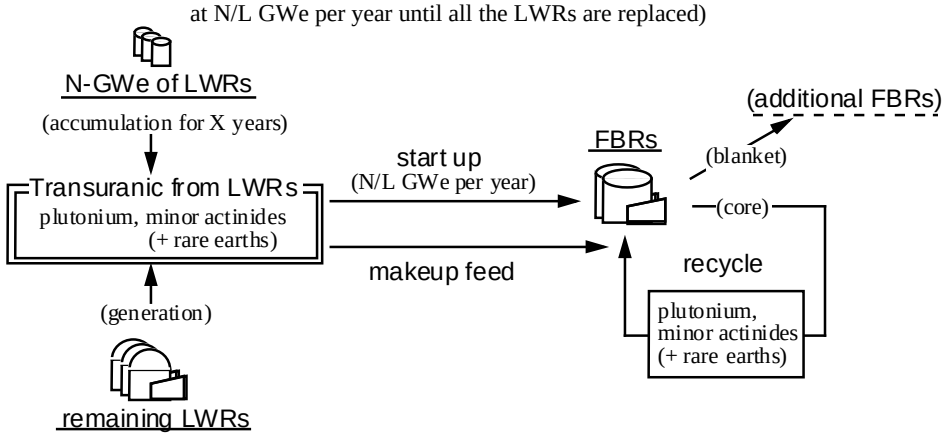
Analytical Mdel

LWR-FBR fuel cycle

Figure 1 illustrates the LWR-FBR fuel cycle model considered in this study. It is supposed that a fixed scale of power generation by LWRs is continued for a certain period until the introduction of FBRs begins. The transuranic elements recovered from the spent fuel are accumulated during this period. After this period, FBRs are started up to replace those LWRs that reaches the reactor lifetime, so that the scale of the power generation is maintained. More than 1/L of the LWRs should be replaced per year if the lifetime is L years. The accumulated transuranic from the LWRs are used to both startup and feed the FBRs. The transuranic generated by the remaining LWRs after the beginning of the FBR introduction are also fed to FBRs. If any transuranic from the LWRs remains after L years, when all the LWRs are replaced by the FBRs, it is used to renew earlier FBRs that reach the lifetime.

All the Plutonium and minor actinides discharged from the FBR cores are reprocessed and recycled to the FBR cores. The minor actinides content in the recycled FBR fuel is set to be larger than their fraction in the FBR spent fuel, so that some minor actinides from the LWRs can be accepted. The Plutonium enrichment is determined to satisfy the refuelling interval length and other design specifications for the FBRs. As long as the Plutonium from the LWRs remains, it is used to make up the enrichment of the recycled fuel. The Plutonium bred in the blanket goes out of this fuel cycle model. It is used to build additional new FBRs.

Figure 1. **An LWR-FBR fuel cycle model**
(N-GWe of LWRs are operated for X years before the replacement by FBRs begins.
The reactor lifetime is L years and the replacement is continued
at N/L GWe per year until all the LWRs are replaced)



Composition of the transuranic from LWRs

A large PWR is chosen to represent the LWRs and the averaged discharge burnup is set to be 48 GWd/t or 60GWd/t reflecting the ongoing effort toward higher burnup goals. The refuelling batch number is three for both cases, with the interval of 15 or 18 months respectively. The burnup and following decay calculations are conducted with ORIGEN2 code. The discharged material is reprocessed after 5 years cooling time and all the transuranic are partitioned and recovered. The overall recovery ratio of 99.5% is assumed for all the transuranic elements according to the target value of the pyro-partitioning technology development [3].The decontamination factor of the rare earth fission products is about 10, which means that the partitioned minor actinides are accompanied by about the same amount(weight) of rare-earth fission products. The composition of the transuranic and the rare-earth elements do not change with the pyro-partitioning. Table 1 summarises the obtained transuranic compositions for the period of 5, 30, 60 and 90 years after discharge. The length of the storage period plays a significant role in increasing the fraction of the minor actinides, mainly due to the decay of ^{241}Pu to ^{241}Am.

Table 1. **The composition of the transuranic from the LWRs** (based on the burnup of one ton-U fuel)

1) Burnup = 48GWd/t

decay period (year)	5	30	60	90
Pu amount (kg)	10.9	9.9	9.6	9.5
Pu composition[a] (w/o)	3/53/24/12/7	2/58/27/4/8	2/61/28/1/8	2/61/28/0/9
MA amount (kg)	1.4	2.3	2.6	2.7
MA composition[b] (w/o)	51/30/14/5/0	33/57/8/1/0	32/60/8/0/0	34/58/7/0/0

2) Burnup = 60GWd/t

decay period (year)	5	30	60	90
Pu amount (kg)	11.5	10.4	10.1	9.9
Pu composition[a] (w/o)	4/51/25/12/8	3/56/28/4/9	3/58/29/1/9	2/59/30/0/9
MA amount (kg)	1.8	2.7	3.0	3.1
MA composition[b] (w/o)	55/25/14/5/0	38/52/9/1/0	36/55/8/0/0	38/53/8/0/0

[a]Pu(Plutonium) composition = ^{238}Pu/^{239}Pu/^{240}Pu/^{241}Pu/^{242}Pu.
[b]MA(minor actinides) composition = ^{237}Np/^{241}Am/^{243}Am/^{244}Cm/^{245}Cm.

The metal fuel FBRs and their fuel cycle

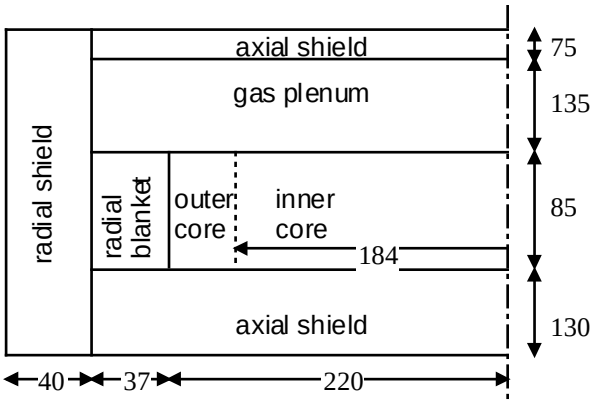
As the model for the commercial FBR, a 1,500MWe metal fuel FBR design is used [4]. The refuelling interval of the core is 18 months and the averaged discharge burnup of 150 GWd/t is achieved. Other major specifications are summarised in Table 2.

Table 2. **The design specifications for the metal fuel FBR**

Item	Specification
Core output	1,500MWe (3,900MWt)
Cycle length	18 months, FPD (4 batches)
Core configuration	homogeneous
Fuel and cladding material	U-Pu-Zr, ferritic stainless
Coolant Temperature (inlet/outlet)	355/510°C
Maximum cladding Temperature	650°C
Maximum linear power	500W/cm
Averaged discharge burnup	150GWd/t

The core has a homogeneous configuration with two Plutonium enrichment regions and radial blankets, as is shown in Figure 2. The Axial blanket is not necessary as long as high breeding performance is not required, since the radial blanket can provide 4-5% fissile gain which is enough to makeup the burnup loss. The neutronic calculations are conducted with a diffusion code that can handle the transuranic burnup and decay chains. Group constants are prepared using JFS3-J32, which is a 70-group constant set for fast reactors [5]. Although the absorption by the rare-earth elements accompanying the minor actinides in the fresh fuel is taken into account, their burning is not considered. A sub-channel code is used for the thermal hydraulic design and the resulting region-wise averaged temperatures are used to calculate the reactivity coefficients.

Figure 2. **Configuration of the 1,500MWe FBR Core. Dimensions are in cm.**



The spent fuel discharged from the core is reprocessed after 3 years cooling time with pyro-reprocessing. The recovered Plutonium and minor actinides are returned to the core. The overall recovery ratio of all the transuranic is 99.5% and the rare-earth fission product decontamination factor is 10. The burnup loss of the Plutonium is made up by the stock Plutonium from the LWRs. Table 3 summarises the core performance characteristics at the equilibrium cycle. Here, the composition after 60 years storage is used for the makeup feed from LWRs. Usually the minor actinides fraction in the FBR discharge is less than 1%. In the case of 'no-MA-makeup' where no minor actinides from the

LWRs are accepted, the content of the recycled minor actinides and rare earth fission products in the fresh fuel come to 0.6% and 0.4% at the equilibrium. In the cases of 2% and 5% minor actinides content, the minor actinides from the LWRs are used to fill up the specified minor actinides content in the fresh fuel. As shown in Table 3, the Plutonium enrichment becomes slightly smaller when the content of the minor actinides is greater. But the safety parameters can be degraded because of the harder spectrum and the fast fission of the minor actinides. Also shown Table 3 are the performance parameters for the case of 'once-through', in which the transuranic from the FBRs are not recycled and the fresh fuel is made only from the material from LWRs. The material requirement for the initial startup is derived from these cases. It should be noted that these cores can achieve a high breeding ratio of >1.35, if upper and lower axial blanket of ~30cm thickness are added. With that high breeding performance, it may be possible to realize a system doubling time of shorter than 30 years.

Table 3. **The FBR feed compositions and core performance parameters at the equilibrium cycle**

	no-MA-makeup [a]	MA-enriched [a]		once-through [a]
MA & RE content [b] (w/o)	0.6/0.4	2.0/1.5	5.0/3.5	2.0/2.0
MA composition [c] (w/o)	14/34/2/30/17/3	23/46/5/13/11/2	28/52/6/6/6/2	32/60/8/0/0/0
Pu enrichment [d] (w/o, inner/outer)	14.8/20.3	14.7/19.7	14.1/19.3	15.0/20.1
Pu composition [e] (w/o)	2/58/30/4/6	3/57/30/4/6	8/54/28/3/7	2/61/28/1/8
Burnup reactivity (%_k/k)	2.0	1.8	1.5	1.9
Internal conversion ratio	0.90	0.90	0.91	0.88
Breeding ratio	1.07	1.07	1.09	1.05
Doppler const. (%Tdk/dT, $x10^{-3}$)	-2.09	-2.20	-2.26	-2.27
Coolant coeff. (¢/°C)	0.254	0.273	0.310	0.279

[a] no-MA-makeup = no minor actinides from the LWRs are accepted.
MA-enriched = minor actinides from the LWRs are accepted according to the fixed content specification.
once-through = no transuranic from the FBRs are recycled.
[b]MA & RE content = weight percent of minor actinides and rare earth fission products in (heavy metal + RE).
[c]MA composition = ^{237}Np/^{241}Am/^{242m}Am/^{243}Am/^{244}Cm/^{245}Cm.
[d]Pu enrichment = weight percent of Plutonium in (heavy metal + RE).
[e]Pu composition = ^{238}Pu/^{239}Pu/^{240}Pu/^{241}Pu/^{242}Pu.

Mass Flow of the LWR-FBR Cycle

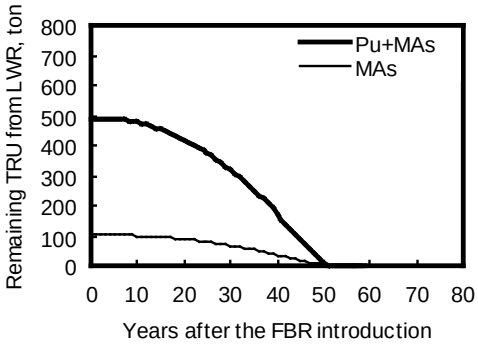
In this section, the calculated results for the mass flow of the LWR-FBR fuel cycle model are described. The scale of power generation can be arbitrary. For this study, 50GWe is chosen for better understanding of the results, which corresponds to the requirement of about 800 ton-Uranium per year when PWRs with the averaged discharge burnup of 48GWd/t are assumed. For simplicity, the transuranic discharged from decommissioned LWRs are not considered. Those recovered from decommissioned FBRs are also not taken into account because they are supposed to go out of this fuel cycle model and be used to build additional new FBRs, together with the Plutonium bred in the blanket.

Figure 3 shows how the Plutonium and minor actinides from LWRs are consumed once the introduction of the FBRs is started after the 50GWe of LWRs are operated for 50 years. The

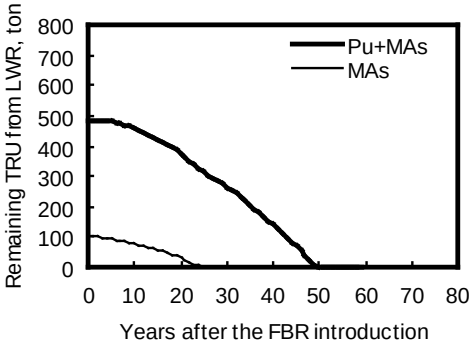
accumulated amount of Plutonium and minor actinides at the year zero are about 400 ton and 100 ton respectively, of which compositions are assumed to be those of the material after 60 years storage. The reactor lifetime is set to be 40 years and, accordingly, 1/40 of the LWRs is replaced by FBRs of which minor actinides content is 2%. The remaining amount of the transuranic from LWRs starts to decrease as the replacement continues, and both Plutonium and minor actinides are finished in 50 years. It means that the transuranic accumulated during the extended LWR era can be consumed within the FBR fuel cycle of the same scale, though it takes as long a period as the period before the introduction of FBRs.

Figure 3. **The remaining transuranic from the LWRs after the beginning of the FBR introduction**

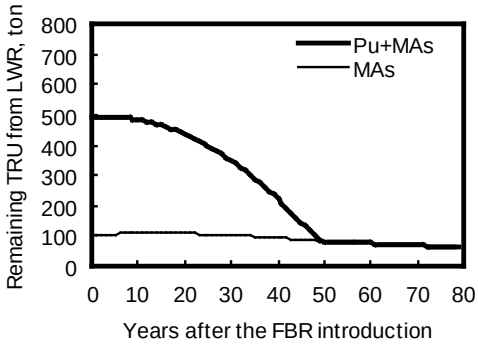
•50GWe LWRs are operated for 50 years before the FBR introduction.
•The LWR discharge burnup is 48GWd/t.
•The reactor lifetime is 40-years, and 1/40 of the LWRs are replaced per year.



(a) The minor actinides content of the FBR fuel is 2%.



(b) The minor actinides content of the FBRs is 5%.

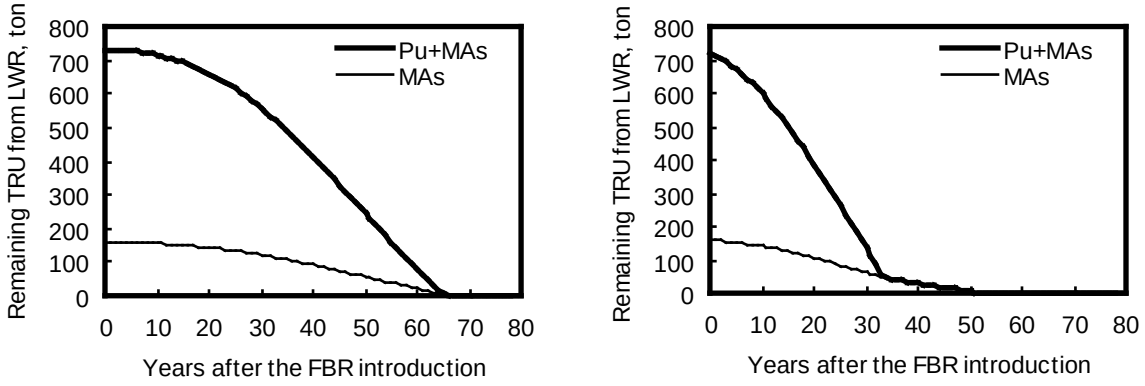


(c) The minor actinides content of the FBRs is 1%.

The changes of the remaining transuranic amount calculated for the cases in which minor actinides contents are 5% and 1% are compared in Figure 3(b) and 3(c). The minor actinides are consumed much faster in the former case, as is shown in Figure 3(b). But it appears not necessary to go up to 5% content considering that it still takes about 50 years to consume all the Plutonium. In the latter case, Figure 3(c), minor actinides can not be consumed as fast as Plutonium and it takes very long to finish all the minor actinides after the Plutonium is finished. It would be needed to utilise the additional FBRs that are started up with the extra Plutonium bred in the blanket.

Figure 4**. The remaining transuranic from the LWRs: the impact of a longer accumulation period**

•50GWe LWRs are operated for 75 years before the FBR introduction.
•The LWR discharge burnup is 48GWd/t.
•The minor actinides content of the FBR fuel is 2 weight percent.



(a) 1/40 of the LWRs are replaced per year. (b) 1/20 of the LWRs are replaced per year.

Figure 4(a) shows the result calculated for the case in which the period of the LWRs operation before the FBR introduction is extended to 75 years. The composition of the accumulated transuranic is assumed to be that of the material after 90 years storage. The content of the minor actinides in the FBR fuel is 2%. The consumption takes longer than that of the cases shown in Figure 3, because of the larger initial amount. It takes about 65 years to finish both the Plutonium and minor actinides. When the rate of the replacement is doubled, the Plutonium consumption is more accelerated than the minor actinides consumption and some amount of the minor actinides is left after the Plutonium is finished, as is shown in Figure 4(b). But this can be easily consumed in the operating FBRs and finished before 50 years.

Figure 5 shows the results for the cases where the burnup of the LWRs is raised to 60GWd/t and the period before the FBR introduction is either (a) 50 years or (b) 75 years. The content of the minor actinides is 2%. The scale of the heavy metal mass flow is reduced to 640 ton-U/y, reflecting the higher burnup. The minor actinides tend to remain slightly longer than the Plutonium, due to the larger fraction in the LWR spent fuel. But the overall consumption of the accumulated transuranic is quicker than the cases of Figure 3 and 4 owing to the smaller initial amount.

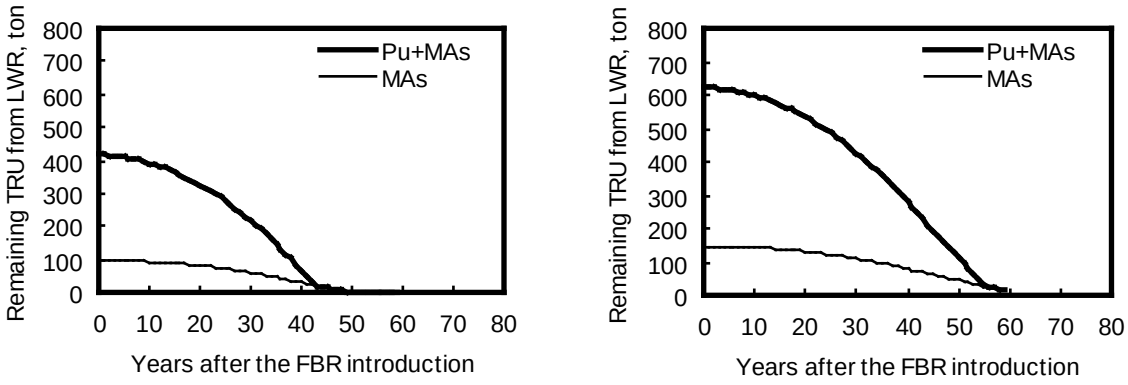
These results indicate that further delay of the FBR introduction causes no significant problem in terms of the consumption of the accumulated transuranic.

Figure 5. **The remaining transuranic from the LWRs. The impact of the LWR burnup**

•The LWR discharge burnup is 60GWd/t.

• 1/40 of the 50GWe of LWRs are replaced per year.

•The minor actinides content of the FBR fuel is 2 weight percent.



(a) The period before the FBR introduction = 50 years

(b) The period before the FBR introduction = 75 years.

Conclusion

The mass flow of Plutonium and minor actinides is calculated for a future LWR-FBR fuel cycle model, in which a certain scale of power generation by LWRs is continued for 50 to 75 years before the replacement by FBRs begins. The averaged discharge burnup of the LWR spent fuel is set to be 48 GWd/t or higher considering the current goals for high burnup. It is assumed that all the Plutonium and minor actinides recovered from LWRs are kept and used to startup and feed metal fuel commercial FBRs. All the heavy metals discharged from the FBR cores are reprocessed and recycled.

The results show that the Plutonium and minor actinides from LWRs can be consistently consumed without further accumulation, by establishing the same scale of FBR power generation and its fuel cycle. The optimum content of the minor actinides in the standard FBR fuel is about 2 weight percents. The consumption may take longer when the period before the FBR introduction is longer, due to the increased amount. But still it is possible to consume the accumulated transuranic by simply introducing FBRs. The best way to consume the transuranic of the LWR origin is the recycle into the FBRs. It can be concluded that the extended LWR era causes no significant problem in terms of the consumption of the accumulated transuranic, if the FBR era is to come in the future.

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A COMPARISON OF SOME NEUTRONICS CHARACTERISTICS OF CRITICAL REACTORS AND ACCELERATOR DRIVEN SUBCRITICAL SYSTEMS

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Abstract

After a general introduction of characteristics of possible critical reactor systems (CRS) and accelerator driven systems (ADS) it is proposed only to compare similar systems, based on the same design principles. For fast neutron spectrum designs of CRS and ADS, comparisons concerning applied calculational procedures, flux and power distributions, power level control, burnup behaviour, incineration potential, aspects of dynamics and accident analysis and possible future R&D-work are discussed. Although ADS may have important advantages, especially related to safety aspects, it is not yet clear that during accidents with heat decay removal failures, fuel melting and fuel slumping with the potential of recriticality configurations may be avoided. If such recriticality cannot be excluded, reconsideration of thermal ADS may be a suitable solution.

Introduction

In a critical reactor system (CRS) a neutron chain reaction is kept alive due to the balance of neutron productions and neutron losses. Due to the fact that a small fraction of the produced neutrons is not emitted as prompt neutrons immediately after the fission events but appears after characteristic delay times due to the decay of associated precursors, critical systems may be controlled fairly easily, e.g. for the adjustment of the required power level. Many different designs for CRS have been proposed and realised with very different characteristics. Examples of actual designs for power reactors are: light water reactors with boiling or pressurised coolant (BWR, PWR), heavy water moderated reactors (CANDU), gas-cooled reactors (AGR, HTGR), liquid metal cooled fast reactors (mainly sodium cooled). Molten salt reactors have been investigated since the early fifties.

Accelerator driven subcritical reactor systems (ADS) also have already been proposed in the fifties. A revival of these ideas was initiated by considerations at LANL by Bowman and co-workers for the incineration of long-lived wastes from nuclear applications and by the proposals of Rubbia and co-workers for a so-called energy amplifier and for plants dedicated to the incineration of nuclear wastes e.g. arising from conventional LWRs. Further, in a number of countries like USA, Japan, France, Spain, Italy and others, dedicated projects are in progress for the realisation of such systems. These ADS concepts also show different design features, varying from molten salt to conventional lumped fuel systems. The power level in an ADS is proportional to the applied accelerator currents and depends strongly on the level of subcriticality and on the position of the external sources. In the present paper a comparison of important characteristics of CRS and ADS will be given for comparable designs of the reactor components of the systems.

Main properties of possible designs of CRS and ADS

The characteristics of CRS are well known and are analysed in many publications. In reference [1] an overview for modern reactors may be found. Reference [2] gives more detailed information for fast reactors. Concerning molten salt systems, an interesting review may be found in reference [3]. A comprehensive overview of ADS activities is collected in the IAEA status report [4].

For the comparison of CRS and ADS the most interesting topics are safety and reliability. Economic aspects may be of interest too, but they will not be discussed here. Although all nuclear reactors may claim to be safe, not all reactor concepts have the same **safety characteristics**. The most important safety related issue is the possibility of supercritical configurations of the reactor system with the potential of energetic reactivity accidents leading to core destruction and radioactivity releases. The probability of building critical masses during core damaging accidents also is an important question in this context. Thermal reactors with low enriched fuel are favourable, compared to fast reactors with higher enrichments. Another important property of a reactor system is its dynamic behaviour after disturbances in the nominal conditions. Important parameters for the dynamic reactor behaviour are reactivity coefficients for deviations from the nominal state like coolant density reactivity coefficient, fuel temperature coefficient etc. and neutron lifetime and delayed neutron fraction in the system. Thermal systems usually have positive **coolant density reactivity coefficients**, whereas fast systems have a tendency to have negative ones. A negative coolant density reactivity coefficient means that a decrease of the coolant density leads to a positive reactivity effect. Positive reactivity effects lead to increased power density, increased coolant temperature and decreased coolant density with positive reactivity feedback. In such systems other mechanisms, like fuel temperature coefficients, are required to avoid the reactivity increase. Positive

coolant density reactivity coefficients are highly desirable in CRS. In ADS the level of subcriticality may avoid problems with negative coolant density reactivities. The main components of the **fuel temperature reactivity coefficient** are the temperature dependent reaction rates in the resonance region of the heavy isotopes (Doppler effect). The main contributor to the negative Doppler effect is usually the capture in ^{238}U or ^{232}Th. Other isotopes with significant contributions may be absorber fuel and structural materials. The compensating effects of fission and capture contributions of fissile isotopes usually lead to small net results. The **mean neutron lifetime** decreases with spectrum hardening and may vary from $2.5\ 10^{-5}$ seconds in PWR to $4.5\ 10^{-7}$ seconds in FBR [7]. Larger mean neutron lifetimes are favourable for the reactor dynamics behaviour. The **delayed neutron fraction** in reactor cores depends on the composition of the fuel isotopes. In most cases, delayed neutron fractions from fast fissions (e.g. in ^{232}Th and ^{238}U) are considerably larger compared to the effects for thermal fissions. Also a tendency to smaller values for inceasing charge numbers of the heavy isotopes may be observed [5]. The mean fraction of the delayed neutrons is associated with the value of 1 dollar of reactivity for dynamics behaviour.

The **reliability** of a reactor system is closely connected to the complexity of the technical realisation and to the available experience with such a system. Water cooled reactors have proven reliability, despite the high pressures, especially in PWRs. Heavy metal cooled reactors also have demonstrated proven technology, but with less practical experience. Molten salt technology, as a potential candidate for ADS, has been investigated extensively in the past, but no realised projects exist at present. For ADS the coupling of a reliable powerful accelerator with a suitable subcritical reactor system is a new feature for which until now no practical experience exists. Recent observations with running accelerators indicate problems with disruptions of the proton beams with possible consequences for the residence time of some essential components of the subcritical core [6].

Because the designs of both CRS and ADS may vary significantly, a **comparison of such systems only seems meaningful for comparable proposals, based on the same design principles**. The present discussions and proposals for future ADS concepts concentrate on systems with fast neutrons. Such systems have favourable fission- to capture-rate ratios for most heavy isotopes, being an advantage with respect to actinide production and actinide transmutation. In the following sections a comparison of important characteristics of CRS and ADS with fast neutrons in a reactor core with lumped fuel (fuel-bundles) will be discussed.

Comparison of similar CRS and ADS

The proposal of Rubbia and co-workers for a conceptual design of a fast neutron operated high power energy amplifier [7] lead to a number of related studies. In reference [7] the subcritical reactor core is placed in a huge lead-filled vessel with natural convection for the heat removal. The core consists of hexagonal fuel assemblies of fuel rods with two different lattices in inner and outer zones. The 1 GeV 12.5 mA proton current comes from a dedicated cyclotron accelerator and hits a spallation target near the centre of this core. In 1996 the IAEA started a benchmark investigation for studying the long time behaviour of an ADS with Th/^{233}U fuel as specified in the Rubbia-proposal and with a somewhat simplified geometry model [8]. At FZK own investigations for an ADS similar to the Rubbia-proposal, are in progress, both for an energy amplifier (EA) and for Pu and minor actinide (MA) incineration. First analyses with tools available at FZK showed possible problems for the technical realisation of this concept. Some of these problems are strongly related to the heat production and removal in the core. The investigations for the FZK-proposal will form the basis for the following comparisons. Alternatively to the lead coolant in ADS and CRS, sodium coolant also will be considered. Mainly neutron physics aspects will be discussed.

Applied calculational procedures

For a specific reactor system the applied procedures for neutronic calculations of CRS and ADS are similar. Most codes for the solution of the neutron transport equations for CRS allow the treatment of external neutron sources. In those cases it is convenient to apply an additional code for the determination of the neutron source, produced by a proton beam hitting a spallation target. At present, most of the calculational procedures for ADS investigations apply below ≈20 MeV codes which are also in use for CRS calculations. These codes may be Monte Carlo type like MCNP or deterministic like S_n transport or diffusion solutions. The development of a unified MCNP-based Monte Carlo code for ADS investigations, including depletion, is in progress [9].

Usually the neutronic ADS calculations at FZK are performed within the code system KAPROS in combination with standard stand-alone codes like MCNP and DANTSYS. KAPROS has been developed for the calculation of fast reactor systems and has been supplemented with modules for the calculation of thermal and epithermal reactors. The subsystem KARBUS has been qualified for advanced depletion calculations for thermal, epithermal and fast reactor systems, see also reference [10]. Most of the applied multigroup libraries have an upper energy boundary of 10 MeV. The development of a new library with 75 energy groups up to 50 MeV is presently in progress. For the spallation calculations the LAHET code system LCS and the Jülich-version of HETC have been used. In reference [11] more information is given about these calculational procedures.

Flux and power distributions

A practical nuclear reactor core design needs a power density distribution with acceptable peaking factors. The power shape in a nuclear core is determined by the space-dependent fission rates: neutron flux times fission cross section. Whereas the radial and axial flux shapes in thermal CRS are quite flat, the gradient at the core boundary in fast CRS is steeper. In order to obtain a flatter radial power shape, in fast CRS usually two or three radial fissile enrichment levels are applied. In ADS the power shape seems to be much more problematical. Already in reference [7] it was pointed out, that in a subcritical device far enough from criticality (K_{eff}≈0.95) the neutron flux distribution has an exponential slope towards a neutron source in the centre, compared to a cosine- or Bessel-function shape towards the centre of a cylindical critical reactor. The consequences of this phenomena could be observed in the results of the IAEA ADS benchmark mentioned before [8]. In figure 1 the radial power distributions in 3 ADS with different initial values for subcriticality are shown with the corresponding radial formfactors f_{rad}, varying from 2.5 to 3.8. Such formfactors are not acceptable in practice. At FZK a number of measures have been investigated in order to improve these formfactors [11]. Both multiple enrichment zones and multiple neutron sources in the fast subcritical core allow designs with acceptable power distributions at system start-up. As an example in figure 2 the midplane power distribution in an ADS with uniform fuel lattice and three distributed neutron sources is shown. The overall formfactor for this system is close to f_{tot}≈2.0 and is acceptable for a realistic reactor design.

Figure 1. **Radial power density profiles in an ADS with different criticalities**

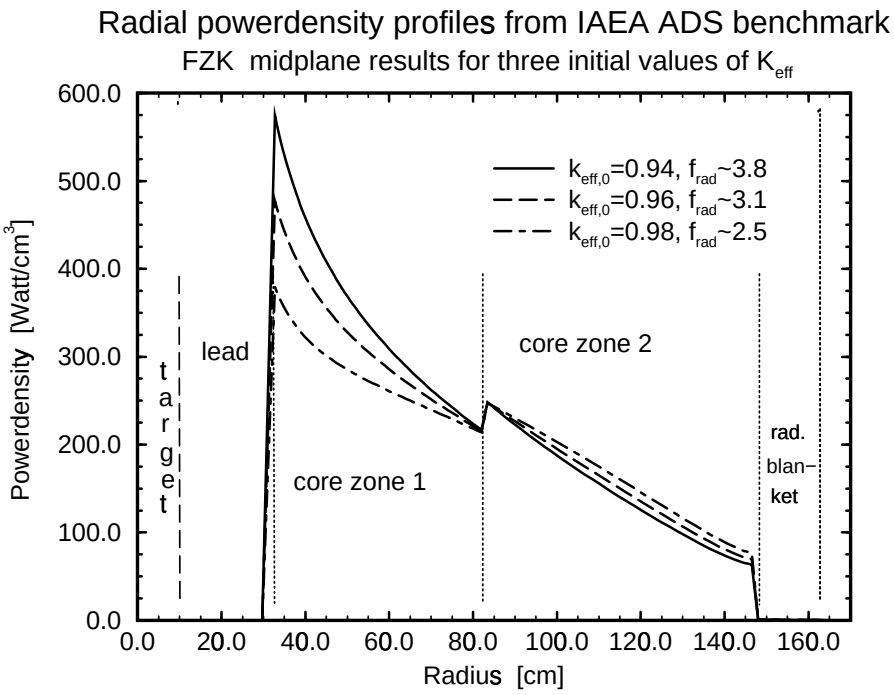
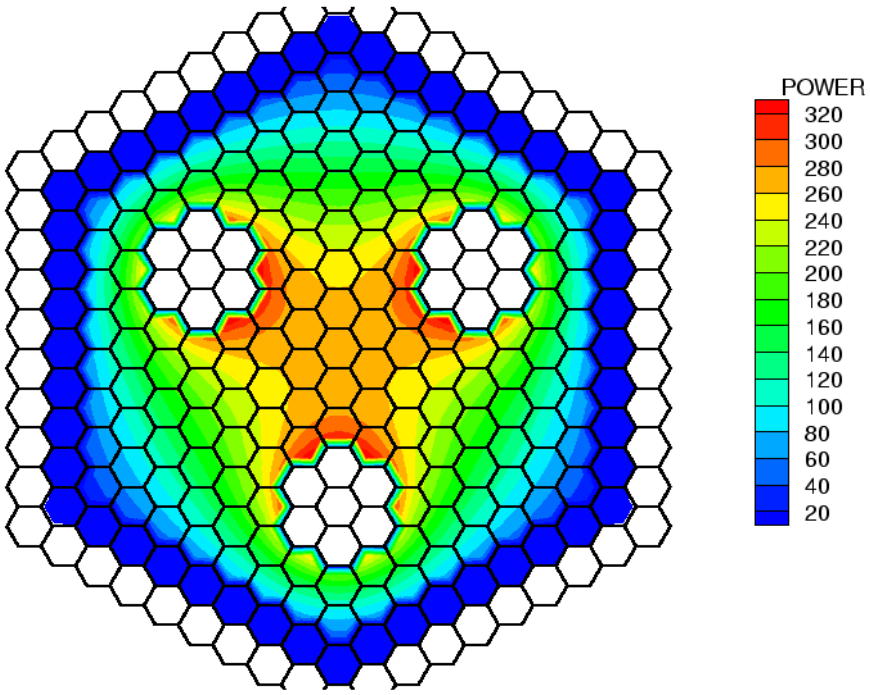


Figure 2. **Midplane power density distribution in an ADS with three neutron sources**



Power level control

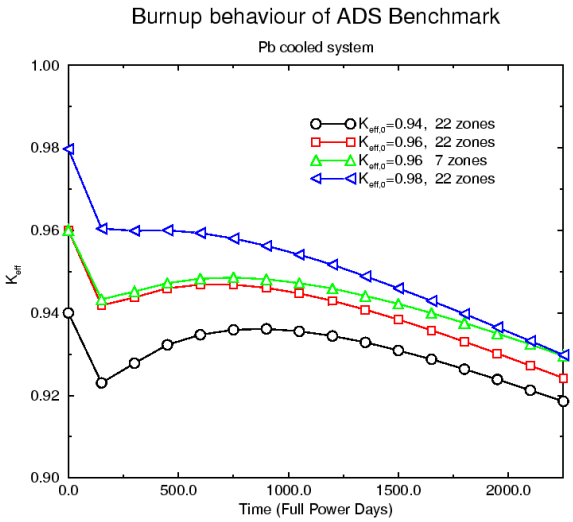
The power level control in CRS and ADS may apply different principles. In both systems the power is determined by the absolute level of the neutron fluxes. The solution of the basic neutron transport calculations for a **CRS** does not provide a value for the absolute level of the fluxes. The principles of critical reactor control, based on delayed neutron fractions, neutron lifetimes and reactivity feedback mechanisms, allow the choice of any arbitrary absolute flux level within the technical constraints of the reactor design. Power level changes in a CRS are initiated by small reactivity disturbations, e.g. by control rod movements or by feedback from changes of thermodynamic properties of the reactor system.

Application of the same calculational tools for an **ADS** introduces an external source in the neutron transport calculation, resulting in absolute fluxes for the ADS case. The power production in the system is determined by the level of subcriticality (internal source) and by the strength and the position of the external neutron source(s). This means, that power level control in an ADS may be realised by changes of the external source strength or of the subcriticality level. Most of the proposals for ADS concepts do not provide active control devices for the subcritical cores and assume proton sources with a rather wide range of operation for the proton currents in order to extract the required amount of power from the core. Such concepts only can be accepted if it can be excluded that an upper level of the subcriticality can never be exceeded, e.g. by reactivity gain due to transmutation processes or decay or due to changes in the reactor configuration.

Burnup behaviour

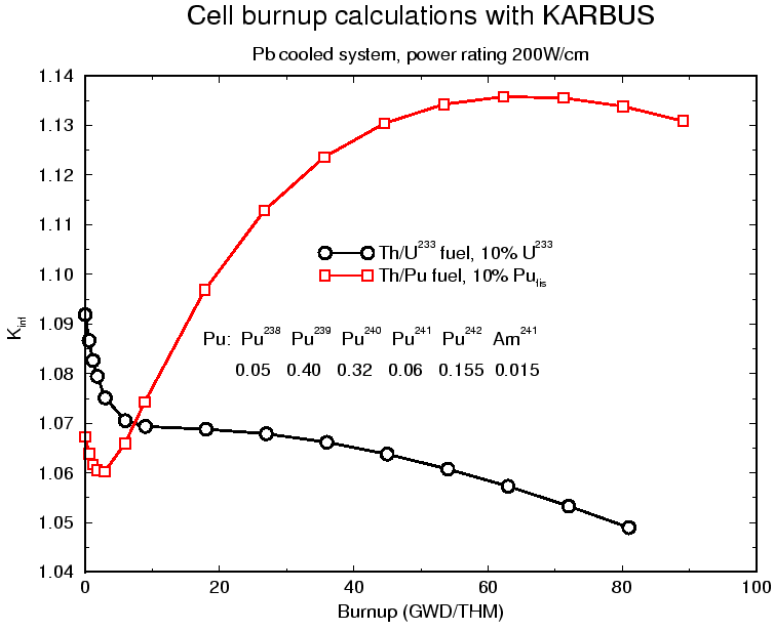
The main processes during fuel burnup are similar in CRS and ADS: changing of the isotopic composition of the fuel and build-up of fission products. Additionally in an ADS also spallation products may play a role, especially in systems with small subcriticality values and corresponding high proton currents. Generally fast neutron spectrum systems have smaller burnup reactivity effects compared to thermal neutron spectrum systems. The main reasons are the lower effective cross sections of the fission products and usually the better conversion of fertile to fissile material in the harder neutron spectrum. The IAEA ADS benchmark in reference [8] was organised to investigate the burnup behaviour of an ADS with Th/^{233}U fuel, see also section 3. Several institutions participated to this benchmark using partly different calculational procedures (codes and data). The FZK results of figure 3 were within the spread of the other contributions. The two curves for initial K_{eff}= 0.96 were obtained with different geometrical models for the burnup zones: 7 zones as specified in the benchmark geometry compared to 22 zones with a finer subdivision in the fuel zones. All curves show a strong reactivity decrease during the first ≈100 full power days. This behaviour is typical for thorium fuel: neutron capture in ^{232}Th leads to the absorber isotope ^{233}Pa being the precursor with half-life ≈28 days of the fissile isotope ^{233}U. After reaching nearly an equilibrium concentration of ^{233}Pa (dependent on the power rating of the fuel), the build-up of ^{233}U may lead to an increase of the reactivity. This reactivity increase is reduced by the absorptions in the fission products. One of the main objectives of the design of Rubbia et. al. was to obtain a flat curve for K_{eff} as a function of burnup. In this way only moderate changes in the proton current are required to maintain a constant system power, without a need for other control mechanisms.

Figure 3. **K_∞ as a function of burnup for the IAEA ADS benmark cases**



Using Pu instead of ^{233}U as fissile material in an ADS, e.g. as a burner of multirecycled Pu with high contents of ^{240}Pu and ^{242}Pu a more complicated burnup behaviour. In figure 4 the results of KARBUS cell burnup calculations are compared for the Th/^{233}U fuel of the IAEA ADS benchmark with 10% ^{233}U content and for a Th/Pu fuel with 10% *Pu* and a Pu composition of 5% ^{238}Pu, 40% ^{239}Pu, 32% ^{240}Pu, 6% ^{241}Pu, 15.5% ^{242}Pu and 1.5% ^{241}Am. We may observe a good agreement between the shapes of the *Th/*^{233}U curve of figure 4 and the IAEA ADS benchmark results of figure 3. However, the Th/Pu fuel in figure 4 shows a strongly deviating burnup behaviour with a strong increase of reactivity with burnup. These preliminary results indicate problems with subcriticality during burnup in such a Th/Pu system and this may lead to a need for active control devices in this ADS, e.g. control rod systems.

Figure 4. **K_∞ as a function of burnup for Th/^{233}U and Th/Pu fuel**



Incineration potential

One of the main objectives in the area of partitioning and transmutation (P&T) research is the incineration of Pu and MA. In the international CAPRA project these issues are studied for Superphenix (SPX) type fast reactor designs. In order to achieve acceptable incineration rates of Pu, as high as possible Pu-fractions in the MOX fuel should be chosen. At present the upper limit for the Pu-fraction seems to be determined by the MOX-solubility during reprocessing at a level of about 45%. These high Pu-fractions lead to a SPX core with highly diluted fuel and large burnup reactivity losses with complications for the reactivity control of this CRS. First exploratory investigations for the incineration of LWR-Pu in an ADS with Th/Pu fuel and sodium coolant show comparable Pu-incineration rates in this not optimised ADS and in the CAPRA reactor; about 70 kg(Pu)/TWe.h ≈ 600 kg(Pu)/GWe.a, see reference 12. The required proton currents in the ADS vary between 18 and 40 mA at 1 GeV. These results indicate, that comparable CRS and ADS designs tend to comparable Pu-incineration potentials. Both systems will have specific advantages and drawbacks. Problems with safety issues in the CRS may be solved for an ADS, but probably at the expense of complex technical ADS designs, especially for the accelerator if high proton currents are required.

One obvious advantage of ADS is the neutron surplus provided by the spallation processes. These (expensive) extra neutrons enable the incineration of materials like fission products or the use of isotopic mixtures with K_∞ values not sufficiently exceeding unity for realistic conventional CRS.

Some aspects of dynamics and accident analysis

Sodium versus lead coolant in fast reactor systems. As explained in section 2, the dynamics behaviour of a reactor system strongly depends on its design, e.g. the choice of coolant and fuel. If we compare a CRS and an ADS with the same coolant and the same type of fuel, the most important difference is the required fissile fraction in the fuel. Depending on the level of subcriticality in the ADS, the enrichment of the ADS may be substantially smaller compared to the CRS. Lower enriched fuel tends to more favourable coolant density and fuel temperature reactivity coefficients and improved dynamics behaviour. Moreover, the number of possible critical masses in the reactor system during a core damaging accident will be smaller, if existent. The choice between sodium and lead for a fast reactor system also may influence the required fissile fuel fraction considerably. As an example, in table 1 preliminary results are shown from an investigation for the potential use of SNR300 fuel assemblies (FA) in a subcritical system. The K_∞ and K_{eff} values have been obtained from eigenvalue calculations using the KAPROS modules DIFF0U and D3E.

Table 1 **Comparison of reactivities with sodium and lead coolant for SNR300 type fuel**

Coolant	K_{oo} DIFFOU	K_{eff} D3E
Sodium	1.6048	0.9659
Lead	1.6192	1.1416

The applied reactor model consists of 169 FA-positions (central position surrounded by 7 rings of FA) with the following loading: in the central 7 positions coolant with the proton source, the positions 8 to 91 (ring 2 to 5) SNR300-core1 FA and the positions 92 to 169 (ring 6 and 7) coolant. The criticality of this model has been investigated both for sodium and lead coolant. The K_∞ values show only small differences for the two coolants. However, in the case of sodium cooling we have an acceptable K_{eff} for the subcritical core, whereas the lead cooling case is far supercritical. This means,

that for this SNR300 fuel in this specific design for a relatively small core with a coolant buffer zone at the outer boundary the enrichment may be substantially smaller with lead cooling, compared to sodium cooling and that lead cooling will have better safety characteristics.

Emergency shutdown in CRS and ADS

Although the energy and heat production in an ADS can be stopped rather easily by interrupting the proton beam, this might not be a decisive advantage compared to CRS. First of all, shutdown and control mechanisms in CRS have demonstrated their extraordinary reliability, based on diversity and redundancy in design and activation. Further, the decay heat removal after shutdown poses an equally severe problem for both CRS and ADS. It has still to be proven that in comparable designs for CRS and ADS, in case of system failures leading to problems with decay heat removal with probably fuel melting and slumping, the consequences for an ADS would be less harmful than for an CRS.

Complications with neutron dynamics calculations of AD.

The neutron dynamic analysis of **near to critical systems** with point or space dependent kinetecs, including perturbation theory, is well understood and many qualified codes are available for different reactor types. These CRS codes are not always applicable to ADS. Some possible complications are:

- For the calculation of feedback reactivities reliable power distributions are required. This means for an ADS that external neutron sources must be taken into account for the determination. Of the neutron flux distributions not all codes have this option.
- For near to critical systems, eigenvalue calculations are convenient and reactivity coefficients are well defined by differences between the eigenvalues. Perturbation theory enables the fast calculation of reactivity coefficients for disturbances from the nominal reactor configuration. In an ADS the multiplicity of the system may be calculated from neutron productions and neutron losses in the system. A straightforward application of perturbation codes is not possible, because these codes are based on real and adjoint fluxes from eigenvalue calculations. So the determination of reactivity coefficients in an ADS may be more complicated compared to CRS.
- For neutron dynamic calculations in near to critical systems usually the following flux synthesis is applied:

$$\Phi(\vec{x},t) = \Psi(\vec{x},t) \cdot P(t)$$

with

$\vec{x}$	variables for space, energy, direction
t	time

A common assumption is that the shape function $\Psi(\vec{x},t)$ varies only slowly with time, whereas the amplitude function P(t) may vary much more rapidly with time. The power density curves of figure 1 in section 32 indicate, that the **power shape in an ADS is quite sensitive to the subcriticality level**. The application of the flux synthesis formulations and simplified treatments such as the quasi-static approaches have still to be qualified for ADS investigations.

Comments addressing fields for further R&D-work for ADS

The needs for R&D-work for ADS are related to several different areas. Some important topics are:

- Accelerator design for high reliable steady state proton currents.
- Spallation target design, including the window.
- Lead coolant technology.
- Thorium fuel cycle technology.
- In the case of MA transmuters, remote handling techniques for fabrication and reprocessing of fuel containing high MA concentrations.
- Calculational tools, e.g. for the improvement of
  - the spallation physics,
  - the coupling of spallation and neutron transport processes including burnup,
  - the neutron physics and thermohydraulic design and
  - the dynamics and safety analysis.

Concerning neutronics the following items may be identified:

- Development of a unified Monte Carlo code, including improved spallation physics coupled with neutron transport and burnup. This activity has been started [9]. Such a code is required for detailed design studies and may be applied for the determination of reference solutions for the qualification of codes with simpler approximations to be used in exploratory investigations.
- Implementation of multi-group libraries with energies above 20 MeV (up to 50..300 MeV), mainly for the deterministic solutions of the neutron transport equations. The upper energy boundary may be determined by sensitivity studies.
- Development of fast nodal neutron transport codes with an adequate external source option, e.g. modification of the HEXNOD code.
- Integration of the spallation product and the fission product treatment. This issue becomes of growing interest and significance with decreasing. core subcriticality and related higher proton currents.
- Formulation of a modified perturbation theory for the calculation of reactivity coefficients for source driven subcritical systems.
- Analysis of the validity of quasi-static approaches for dynamics investigations and possibly development of more advanced methods in case this approximation is not justified and its application is not adequate.

Concluding remarks for future investigations for ADS

In the past years a number of proposals for ADS applications have been presented, varying from an energy amplifier with lumped Th/^{233}U fuel and fast spectrum with lead coolant to fission product and actinide transmuters with fast and thermal spectra. The main argument for these ADS concepts is usually improved safety, compared to CRS. Some ADS objectives hardly may be realised with CRS,

.e.g. incineration of larger amounts of fission products or of heavy fuel isotopes with unfavourable properties for the dynamics behaviour (delayed neutron fractions, coolant density and fuel temperature reactivity coefficients, etc.). A comparison of similar CRS and ADS designs shows an improvement of the safety characteristics of an ADS, caused by the lower required fissile enrichments. However, it is not clear that in fast spectrum ADS recriticality during core damaging accidents can be avoided in all circumstances. In the case core damaging accidents with recriticality cannot be excluded, reconsidering thermal ADS may be a more suitable solution. Finally one has to keep in mind that both **critical and subcritical burner reactors** only may be run if reprocessing plants with adequate capacity for the fuel cycles of interest are realised.

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COMPARISON STUDY OF HYBRID VS CRITICAL SYSTEMS IN POINT KINETICS

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Abstract

An essential motivation for hybrid systems is a potentially high level of intrinsic safety against reactivity accidents. In this respect, it is necessary to assess the behaviour of an Accelerator Driven System during a TOP, LOF or TOC accident.

A comparison between a critical and sub-critical reactor shows a larger sensitivity for the critical system. The ADS has an unquestionable advantage in case of TOP but a less favourable behaviour as for LOFWS type of accidents. However in the ADS cases, the beam could be easily shut off during the transient. Therefore, a part of the R&D effort should be focused on the monitoring and control of power.

Introduction

In France, the Hybrid systems are dedicated to the elimination of minor actinides and long life fission products according to "the France Act of 1991". The main interest of this type of system is a high potential intrinsic safety level as regards reactivity accidents, in spite of a low dollar value and negative thermal reactions.

In this sense it is necessary to assess the behaviour of a sub-critical reactor piloted by a source during transient of power (TOP), flow (LOF) or current (TOC) and to compare the results with those of a critical system.

The presentation is illustrated by using a PHENIX type fast neutron reactor model with or without source.

Critical and sub-critical system kinetics behaviour

Neutronic kinetics neighbouring the criticality

If $k_{eff} \simeq 1$, or furthermore $\rho * (k_{eff} -1)/k_{eff} \simeq 0$, then the neutronic kinetics are ruled by the effective fraction keff for the delayed neutrons. Typically, the T_d doubling time for the neutronic population according to the reactivity of the following values (1 cent = $\beta_{eff}/100$):

Table 1. **Doubling time according to the reactivity**

ρ (cent)	1	5	10	25	50	75
T_d (s)	700	140	52	14	3	0.7

As long as the reactivity remains well below the dollar, T_d is in seconds. The reactor control is therefore possible and the usual safety systems are operative.

Prompt criticality

If the reactivity ρ is superior to β_{eff}, the neutronic population is therefore ruled by the prompt neutrons. Its evolution is exponential with a doubling time inferior to the tenth of second. The reactor becomes uncontrollable and the usual action time of the safety system, ca. a few hundred milliseconds, can turn out to be inefficient to prevent a criticality accident.

Neutronic kinetics of a sub-critical system

If l represents the prompt neutrons lifetime and S the neutron source - inherent, external or in spallation - then the neutronic population is equal to $\eta = S * l/\rho$.

Dividing the ρ reactivity by an f factor or increasing the S source by the same factor leads to a multiplication by f of the neutronic population. This effect is instantaneous for a sub-critical system of several dollars.

For example, starting at - 2 $, a 1$ insertion finally entails a doubling of the power.

This behaviour is fundamentally different from that of a critical system for which + 1 $ leads to prompt criticality. This advantage is essential.

On the contrary, in a critical system the S source being insignificant, a negative insertion of reactivity entails the interruption of the chain reaction, which is not the case for a sub-critical system whose source remains constant.

Thermal feedback effect

Any temperature modification in a reactor entails a core reactivity variation. The Doppler effect, the concentration variations of the material in the core and the differential core-tank-control rods expansion are the main thermal feedbacks influencing the reactivity. These reactions are called feedbacks as they oppose the initial disturbance, thus playing a stabilising role. The main exception to this rule concerns the draining of a core cooled by a liquid metal, as in the fast neutron reactor. In such a case, the draining effect can be highly positive in certain areas of the core and therefore evaluated in dollars.

For a power reactor, due to the sensitivity of the neutronic flux as regards the reactivity, the thermal feedbacks' role is essential both for operation and safety. In the case of Hybrid systems, the situation is different. On the one hand, as their function consists in burning minor actinides, long life fission products and possibly Plutonium, the feedbacks are weak- notably the Doppler effect and non-differential expansion in absence of control rods. On the other, the sensitivity of the neutronic power as regards the reactivity is lower than in the critical system as shown in previous paragraph 'Neutronic kinetics of a sub-critical system'.

Reactivity accidents

Typically accidents can have two origins:

- The ramps change of reactivity or transient of power (TOP).
- The loss of flow (LOF) which can possibly lead to a TOP in case of coolant draining.

For Hybrid systems piloted by an accelerator, the transient of current (TOC) must also be taken into account.

Triggered by the injection of the whole margin of the current of the accelerator required for the reactor operation over the total cycle time, they can cause a power overshoot.

Transient of power (TOP)

Theoretically, that is to say without prejudging the accident triggering physical phenomenon, we study the critical and sub-critical system behaviour as regards the insertion of a non-protected fast ramp change of reactivity.

As a first example, a ramp change of reactivity of 0.55 $/s is inserted in a PHENIX type core until the fuel melting is achieved. Two calculations are performed, one with k_{eff} = 1, the other with k_{eff}= 0.95 + constant external source.

The mean temperature and the power evolutions are given in Table 2. The following results are obtained [1].

Table 2

	Critical reactor k_{eff} = 1	**Sub-critical reactor k_{eff} = 0.95**
Time before melting	2 s	12 s
Inserted reactivity	1.1 $	6.6 $
P/Po	2.2	1.5

A factor of 6 on the time before melting and therefore on the inserted reactivity is observed between the two systems.

As a second TOP example, we can use the study performed by Rief and Takahashi [2]. The considered reactor, which is sodium cooled, has a power of 1 GW. The chosen operating mode is either critical or sub-critical of - 1 $, - 2 $ and - 3 $. A ramp change of reactivity of 170 $/s is injected in 16 ms, by a total of 2.72 $. The calculation results show that in critical mode, the prompt criticality is achieved 6 ms after the beginning of the ramp with the first power peak of 700 GW at 8.5 ms and a second at 500 GW of 13.2 ms whereas in sub-critical mode, the peaks are respectively obtained at 530 GW for - 1/$, a maximum of 6 GW for - 2 $ and 2.2 GW for - 3 $ at 16 ms. Even if for an initial sub-criticality of only - 1 $ the power peak is close to the peaks obtained in critical mode, the total energy released during power excursion is much lower.

The results presented here concern non-protected transitories of power. As long as the system remains sub-critical, the power increase with the reactivity is considerably slower than in critical mode thus giving a much longer time for the safety systems to react. Furthermore, it is interesting to note that it is unquestionably faster, simpler and more reliable to cut the spallation source rather than to drop the safety rods to stop the chain reaction.

Loss of flow (LOF)

Let us consider a decrease, or even a total loss of coolant fluid flow which could lead to the draining of the core by boiling, if the accident is unprotected.

As a first example, a primary flow decrease from 100 % to 60 % in 10 seconds is simulated in a PHENIX 1 type core. Two calculations are performed, one with k_{eff} = 1, the other with k_{eff} = 0.95 + constant external source.

While considering the same set of thermal feedbacks, we note (Figure 4) a smaller neutronic power decrease in sub-critical (a power increase could even be possible for a core without control bars if the reactivity effect of the coolant is superior to the Doppler effects and fuel actual expansion). The clad and coolant overheating is then faster and broader than for a critical reactor. Although the sodium boiling temperature is not reached, a larger number of clad breakage should be expected.

As a second ULOF example, we can use the study performed by Rief, Magill and Wider [3]. The considered system is a sodium cooled 800 MWe reactor with a draining effect of 4 $. The primary pumps slow down on their inertia by a factor 2 per 12 seconds.

After a few tens of seconds in critical mode, the sodium boiling leads to the core draining, thus making the critical reactor prompt, the maximum power peak reaching 1,800 times the initial power. The accident causes a fast core melting of the explosive type.

In the case of a sub-critical reactor of - 10 $ with external source, the sodium boiling starts 12 seconds after the LOF beginning. In 3 seconds, 4 $ are injected by draining. The power reaches a peak of only 1.5 times its initial value but however remains at a sufficient level after the peak to entail a "slow" core fusion.

It should be noted that for a lead cooled reactor, the clad melting takes place before coolant boiling. Therefore the accidental sequence will be different from that of a sodium cooled reactor.

In principle a ULOF is more favourable for a critical reactor as the coolant temperature increase is slower and weaker. Nevertheless, if the boiling occurs, the consequences following the TOP caused by the draining are more serious.

In comparison to a TOP, the time before the LOF accident is much longer as it is estimated in seconds. The safety system non-intervention probability (rod drop, source shutdown, Diesel take-over, etc.) is therefore much smaller.

Transient of current (TOC)

The reactivity evolution during an operating cycle requires a spallation source adjustment and therefore that of the current of the accelerator, in order to maintain the power at a constant level. The accidental injection of the whole margin of the current of the accelerator can therefore cause a power overshoot.

However the ρ_o sub-criticality level chosen for the reactor operation can be sent in order to prevent core damaging. Indeed, if $\Delta\rho_{TOP}$ is the reactivity variation between the beginning and the end of the cycle, then the instantaneous introduction of the whole accelerator current margin aimed at compensating $\Delta\rho_{TOP}$ entails a power variation equal to:

$$\frac{P}{P_0} = 1 + \frac{D\rho_{TOP}}{\rho_0 + \beta}$$

If P_{max} is the maximum acceptable power required for accident detection during a short period of time, then ρ_o (i.e. k_{eff}) can be set so that P always remains inferior to P_{max}.

Conclusion

If the core is correctly dimensioned to remain sub-critical in any circumstance, then hybrid systems present an unquestionable advantage compared to the critical systems as regards the transitories of reactivity. The power and temperature evolutions are sufficiently slow to allow intervention of the safety system.

In case of unprotected loss of flow (ULOF), the power slump is faster and broader for a critical system than for a hybrid. Nevertheless, if coolant fluid boiling occurs, the consequences following the TOP caused by draining are more serious. Finally, it seems simpler, faster and more reliable, at the same detection level, to cut the spallation source rather than drop the safety bars to stop the chain reaction

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LIMITATIONS ON TRANSMUTATION PERFORMANCE DUE TO REACTIVITY CONSTRAINTS IN ACCELERATOR DRIVEN SYSTEMS

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Abstract

An analysis is presented to characterise the reactivity effects in molten lead energy amplifiers. Limitations in the nominal k-eff value to avoid reaching criticality in abnormal conditions impose restrictions to the nominal value of the neutron flux. If those restrictions become very severe because of safety features, the transmutation performance would not be good enough. Very long burnup periods would be needed to reach a certain level of transmutation cleansing.

Introduction

Transmutation of transuranics into shorter-living nuclei is possible by nuclear fission. In fact, it produces a near-term amplification of radioactivity, because fission products convey about ten decays per fission before reaching nuclear stability, and those decays happen in a period shorter than 400 years. On the contrary, a transuranic nucleus will also go through ten decays (in rough numbers) before arriving to a stable isotope of lead, but it will take tens of thousand years to arrive to stability.

A critical point to reduce the TRU inventory is to minimise the capture cross section in relation to the fission cross section. Otherwise, higher-A actinides would appear and it would imply longer decay periods to reach stability. This criterion can be met either with very fast neutron spectra or well moderated spectra. The region of cross section resonance must particularly be avoided.

A second fundamental point in this context is to have a high transmutation rate, and this means a high $\sigma\phi$ value. In thermal reactors, σ is very large, but ϕ is small for a given neutron concentration density (N) because the neutron speed is low. On the contrary, σ is small in fast spectra, but ϕ can be much higher. Anyway, if a substantial fraction of a given inventory has to be transmuted in a given time, the value $\sigma\phi$ must reach a certain value.

The flux level is also connected with the power density in the reactor

$$D(W/m^3) = a\sum \sigma_f^i \phi N_i$$

where the summatory applies to all fissionable isotopes and *a* is a constant for unit change. For the sake of simplicity, let us assume the reactor fuel is mainly made of ^{239}Pu. It could be written

$$D = a\sigma_f^9 \phi N^9$$

Therefore, a limit to D (because of thermal-hyudraulics) is a limit to the concentration of ^{239}Pu (N^9). Of course, if V is the volume of the reactor, it holds $^{239}Pu = V.N^9$. If N^9 has to be reduced because of thermalhydraulics reasons, the reactor volume has to be increased, for a given inventory ^{239}Pu.

In any case, a high flux is needed and this means a high value of the k_{eff} of the subcritical reactor. Otherwise, the spallation source strength would have to be so high that the system would be unfeasible.

On the other hand, the reactor should not attain criticality in any condition, even under accidental circumstances. This means that a large enough margin of reactivity has to be accepted in the nominal design, to have room enough to accommodate the reactivity effect of any abnormal performance.

In the following, an analysis is made on the general features of molten-lead energy amplifiers (MLEA) [1-3], starting from an analytical approach to characterise in a systematic way the main reactivity effects.

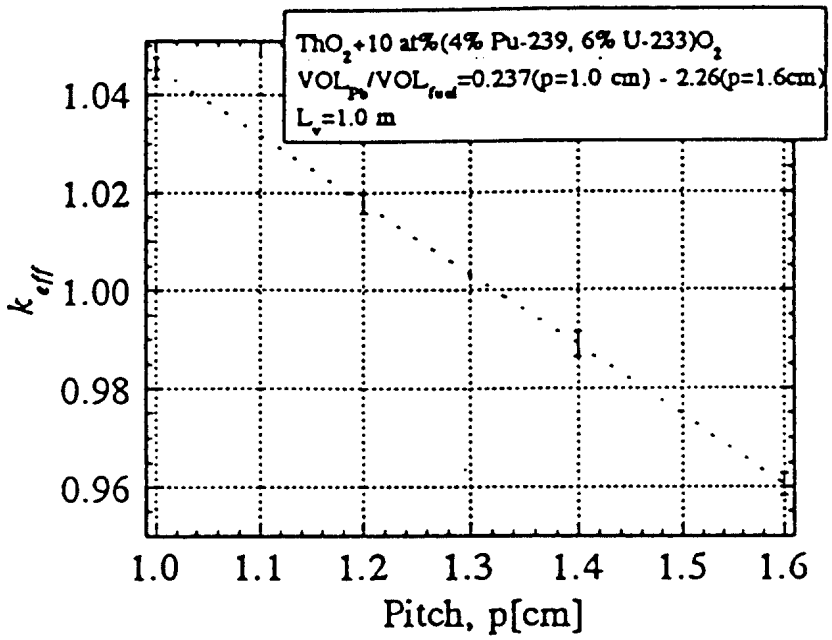
Reactivity variations in a MLEA

Figure 1 presents the evolution of k_{eff} (in a lattice cell calculation) as the cell pitch changes. It must be noted that this calculation implies an infinite array of cells in the horizontal plane. In a realistic design, the fuel mass must be kept constant when changing the geometry, which means that a compacted reactor will have a smaller radius and therefore higher leakage probability.

Compactation simulations must also take into account the accidental filling of the central part of the reactor (where the spallation source is placed) by fuel pins displaced from the periphery inwards. As an example to illustrate this type of simulations, two bare cylindrical reactors have been calculated. The first was made of a homogeneous mixture of lead and fuel (10% ^{233}U) with a radius of 1.67 m and 1.5 m high. The k_{eff} in this case was 0.934. The second cylinder was of the same size as the previous one, but the central axial part (with a radius of 0.4 m) was only made of lead. Its k_{eff} was 0.909. If we take as reference reactor for a MLEA this reactor with k_{eff} = 0.909, it can be seen that the reactor will suffer a 3% increase in reactivity if the central part is also filled with fuel pins.

Another source of reactivity effects is the change in lead density. This is a similar problem to the one found in sodium-reactors and in over-moderated thermal reactors (as happened in the Chernobyl accident [4]. Lead neutronic properties are different from those of sodium. In fact, lead reactor spectrum is very bad for the production of higher actinides because capture to fission cross sections ratio is very small for any nuclei, as compared to the same ratios in a sodium reactor.

Figure 1 **k_{eff} vs cell pitch for a given fuel (4% ^{239}Pu, 6% ^{233}U, fuel pin radius = 0.5 cm)**



The variation of k_{eff} vs lead density is depicted in figure 2. It corresponds to a lattice cell calculations. K_{eff} remains between 0.92 and 0.96, with 0.96 at nominal density. It is important to note that the void coefficient is negative in this case, where the neutron leakage is high because we did not include in the simulation a lead reflector that can eventually remain at nominal density.

A better characterisation was analysed for selected void geometries, described as follows. A cell calculation with 2 m total height, 1.6 cm pitch and 4% ^{239}Pu plus 6% ^{233}U fissile contents was performed for four different lead distributions, which are described in the following lines, including the resulting k_{eff}:

- lead filled at reference density (10 g/cm^3) k_{eff} = 0.962
- total lead void, k_{eff} = 0.929

- central lead void (The lattice cell only simulates the upper half of the pin for symmetry reasons. In this hypothetical case, the lower half of the cell is at 0 g/cm^3 of lead, while the upper half of the lattice cell is at 10 g/cm^3) k_{eff} = 1.00
- outer void (the upper half of the lattice cell is at 0 g/cm^3 and the lower part is at 10 g/cm^3) k_{eff} = 0.917.

The foregoing results are very important because they point out that central voids produces higher reactivity effects. This is due to the harder spectrum created in the central core, where fission probability increases. This is also due to the reflector effect of the peripheral lead. On the contrary, when the void occupies outer regions, reactivity decreases because leakage of very fast neutrons is enhanced. The former physical phenomenon can further be illustrated with the calculation of another accidental scenario. In this case, it is presumed that the cladding material melts down and it goes up because of its buoyancy. Both the plenum (inside the cladding) and the cladding itself are replaced by lead. After that, it is presumed that lead density goes down (because of boiling bubbles) from nominal density (10.6 g/cm^3) to 0.0 g/cm^3. In table 1, a set of calculations of this type is presented. The reference case is a cylindrical reactor 1.2 m high plus 1.5 m plenum, with a radius of 123.6 cm and a fuel composition 86% ^{232}Th and 14% ^{239}Pu. The volume ratio Pb/fuel is 3.23 in this case. The rest of the cases in table I are denoted by the lead density in the reactor region. Outside the reactor, the lead density remains always 10.6 g/cm^3 in the whole vessel.

Figure 2 **k_{eff} vs Lead density in the standard cell calculation (4% ^{239}Pu, 6% ^{233}U)**

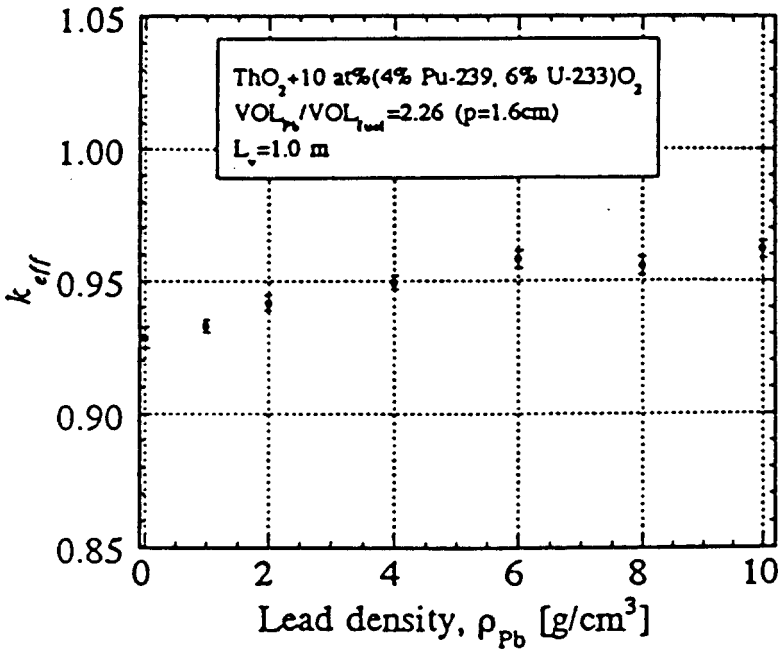


Table 1. **Reactivity injections in the case of clad melting and partial or total lead boiling**

Case (lead density g/cm^3)	k_{eff}	$\frac{\Delta k}{k}$	$\left(\frac{Fis}{Abs}\right)_f$	Abs-fuel Total Abs
Reference	0.913	0.0	0.395	0.955
10.6	0.933	0.022	0.400	0.968
9.54	0.937	0.026	0.410	0.972
6.35	0.948	0.038	0.420	0.977
2.65	0.962	0.054	0.430	0.990
1.06	0.965	0.057	0.435	0.995
0.0	0.971	0.063	0.440	1.000

Additionally, catastrophic accidents could be simulated to some extent by presuming clad melting and fuel desegregation. The fuel can go up (if it is UO_2-PuO_2)within the lead pool, and will expand laterally and shrink vertically until occupying the upper part of the vessel (below the molten clad). The results of those calculations are reported in table 2, starting from the same case of reference as table 1. Again, some increase in reactivity is found, but the lateral expansion of the (very hot) fuel prevents the reactor to achieve criticality, and the final state is highly subcritical. Of course, radioactivity confinement and emergency cooling would be the main problem in this case, but it is important to assess that the final state can not be critical.

All these calculations have been done with the LAHET [5] and MCNP [6] codes. Neutron cross-sections have been taken from ENDF B/IV [7] and ENDL [8] libraries. The JENDL 3.2 set [9] has also been used.

Table 2. **Reactivity effects due to fuel disassembly**

Case	Radius (cm)	Height (cm)	% of original lead	k_{eff}
Reference	123.6	120	100	0.913
I	126.3	100	85	0.946
II	140.1	70	70	0.947
III	149.6	25	10	0.954
IV	342.0	3.6	0	0.215

Summary

Reactivity analysis on potential changes of the reactor geometry and material composition will play an important role in the Safety Report of MLEA [10]. The need to keep subcriticality under any abnormal event that could be conceived from a practical point of view, imposes the need to have a large enough subcriticality margin. This means that the neutron flux will have a limited value and the transmutation rate will also be limited. In the Introduction, it was underlined the need of very high fluxes in order to get rid of the main TRU in a technological time span. In the Appendix, a theoretical analysis is presented to point out the cleansing levels that could be achieved in a MLEA, depending on the flux level. It is seen that ^{239}Pu transmutation is possible down to very low residual fractions. On the contrary, it is difficult to get low residual fractions (below 10%) for ^{240}Pu, which is mainly transmuted by conversion to ^{241}Pu. In any case, the need of a high flux level is paramount and it implies the need for k_{eff} as close to 1 as possible. This trend will be limited mainly by reactivity analysis related to accidental conditions.

Appendix

A theoretical study on Pu isotope transmutation

In the inventory of TRU and their associated radioactive progenie from spent nuclear fuel, the most important isotopes are ^{239}Pu, ^{240}Pu and ^{241}Pu. The first one is a very good fissile isotope, specially in fast spectrum. The last one is still better because it presents a very high σ_f and very low σ_c. If it is transmuted without allowing a long decay span, the amount of ^{241}Am built-up remains small, and the residual radiotoxicity of TRU can be highly decreased by eliminating the afro mentioned Pu isotopes. ^{240}Pu is not a good fissile material (and it is still worse for thermal spectra) but in fast spectrum it can be eliminated either directly or by conversion to ^{241}Pu.

In the following, the inherent burnup features of Pu isotopes transmutation are studied in an analytical approach. It is presumed that a subcritical reactor (in an ADS) provides for a flux ϕ and creates a spectrum in which the microscopic cross section for these isotopes are known. In our case, we take the following values typical of a MLEA [1].

$$\begin{array}{lll} ^{239}\text{Pu} & \sigma_a = 2.21 & \sigma_f = 1.75 \\ ^{240}\text{Pu} & \sigma_a = 0.82 & \sigma_f = 0.35 \\ ^{241}\text{Pu} & \sigma_a = 2.43 & \sigma_f = 2.43 \end{array} \qquad \text{(A.1)}$$

Conversion of ^{241}Pu into ^{242}Pu is not taken into account in the model, because it accounts for much less than 1% of the radiotoxicity. It is also assumed that the reactor is loaded (in any cycle) with the Pu discharged from LWR, which corresponds to the following inventory (per ton of initial Uranium loaded in the LWR)

$$\text{Pu}^9(0) = 5.19 \text{ kg} \qquad \text{Pu}^0(0) = 2.17 \text{ kg} \qquad \text{Pu}^1(0) = 1.03 \text{ kg} \qquad \text{(A.2)}$$

We use these figures as starting point. After each burnup cycle, the total amount of eliminated Pu is computed, and the same mass is reloaded (with the isotopic composition given above). The governing equations are:

$$\frac{d\text{Pu}^9}{dt} = -\sigma_a^9 \phi \text{Pu}^9 \qquad \text{(A.3)}$$

$$\frac{d\text{Pu}^0}{dt} = -\sigma_a^0 \phi \text{Pu}^0 + \sigma_c^9 \phi \text{Pu}^9 \qquad \text{(A.4)}$$

$$\frac{d\text{Pu}^1}{dt} = -\sigma_a^1 \phi \text{Pu}^1 + \sigma_c^0 \phi \text{Pu}^0 \qquad \text{(A.5)}$$

that can be solved analytically to find

$$\text{Pu}^9(t) = \text{Pu}^9(0) e^{-\sigma_a^9 \phi t} \qquad \text{(A.6)}$$

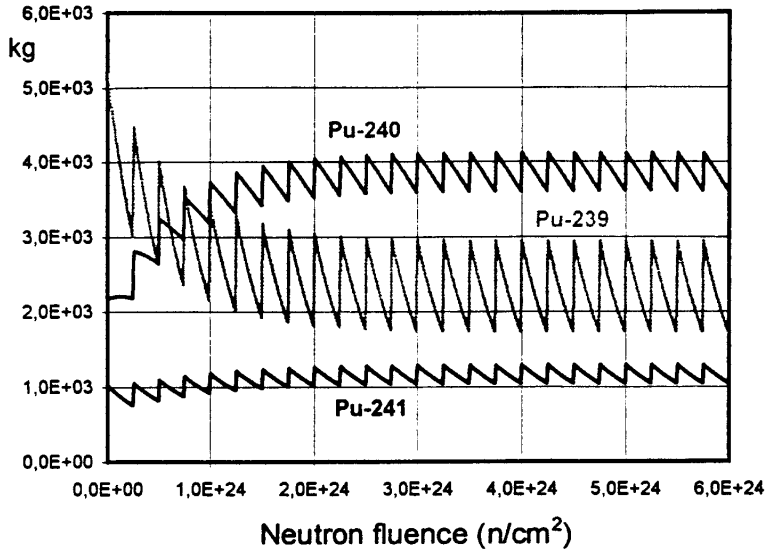
$$Pu^0(t) = Pu^0(0)e^{-\sigma_a^0\phi t} + Pu^9(0)\frac{\sigma_c^9}{\sigma_a^9 - \sigma_a^0}\left[e^{-\sigma_a^0\phi t} - e^{-\sigma_a^9\phi t}\right] \tag{A.7}$$

$$Pu^1(t) = Pu^1(0)e^{-\sigma_a^1\phi t} + Pu^0(0)\frac{\sigma_c^0}{\sigma_a^1 - \sigma_c^0}\left[e^{-\sigma_a^0\phi t} - e^{-\sigma_a^1\phi}\right] +$$
$$+ Pu^9(0)\sigma_c^9\sigma_c^0\left[\frac{e^{-\sigma_a^9\phi t}}{(\sigma_a^0 - \sigma_a^9)(\sigma_a^1 - \sigma_a^9)} + \frac{e^{-\sigma_a^0\phi t}}{(\sigma_a^9 - \sigma_a^0)(\sigma_a^1 - \sigma_a^0)} + \frac{e^{-\sigma_a^1\phi t}}{(\sigma_a^9 - \sigma_a^1)(\sigma_a^0 - \sigma_a^1)}\right] \tag{A.8}$$

In figure A.1, the evolution of the inventory is given for 24 cycles of 2.5×10^{23} n/cm^2 fluence every one. It is presumed that the microscopic cross sections do not change along the burnup, which is a hypotheses that can not be admitted for detailed calculations [11,12] of a given reactor in a given cycle, but is acceptable for an analytical study. It is observed in figure A.1 that an equilibrium cycle is achieved after 12 cycles. After that, the material unloaded from the ADS reactor has a composition given by the following fractions

$$^{239}Pu = 0.2735 \qquad ^{240}Pu = 0.5625 \qquad ^{241}Pu = 0.164 \tag{A.9}$$

Figure A.1 **Evolution of the Pu inventory in successive cycles of transmutations using LWR spent fuel (see text)**



The residual fraction of each isotope is given in figure A.2. It is defined as the inventory of an isotope still existing in the ADS divided by the total amount that has been loaded in the previous cycles. Figure A.3 depicts an integral view of the accumulated inventory that was loaded in the ADS along the successive cycles and the total amount that was transmuted. The figures clearly point out an almost asymptotic behaviour in the Pu-isotopes elimination, if the previous scheme is adopted. Of course, the ADS fuel becomes more and more ^{240}Pu dominated, which enables us to reconsider a second pattern of recycling. Instead of keeping the isotopic composition of the LWR-unloaded Pu, after reaching the equilibrium cycle in ADS, the successive cycles must be loaded with the isotopic

composition of the Pu discharged from the equilibrium cycle (i.e., there is a second phase that can be called ADS recycling). The isotopic composition was given before in Eq. (A.9)

Figure A.2 **Residual fraction of Pu isotopes for the case given in A.1**

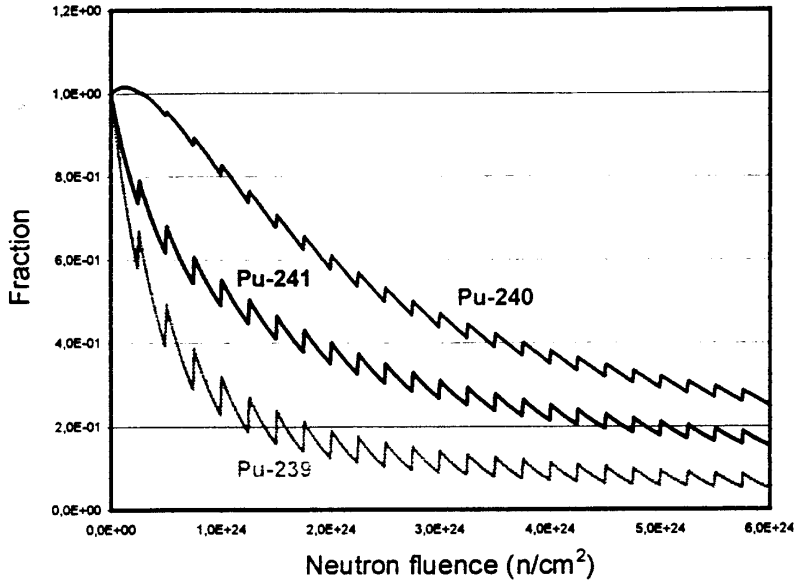


Figure A.4 shows the evolution of the residual fraction of the Pu-isotopes, as defined for figure A.2. Here, a third phase was set up after cycle 24, once the new isotopic composition (of Eq.(A.9)) reached again the equilibrium cycle. In this case, the isotopic composition of the discharged Pu is

$$^{239}\text{Pu} = 0.0962 \qquad ^{240}\text{Pu} = 0.7054 \qquad ^{241}\text{Pu} = 0.1984 \tag{A.10}$$

Of course, the very small amount of Pu-odd isotopes poses a problem about the quality of the fuel in order to get a high-enough k_{eff}. This problem disappears if it is considered that the ADS neutronics is mainly boosted by ^{233}U (albeit this isotopes poses a new problem, related to its own radiotoxicity along a very long time span, because its half-life is ~ 1.6×10^5 years).

Figure A.4 depicts that the residual fraction of ^{239}Pu reaches very low values, of the order of 0.1%, while ^{240}Pu arrives to ~ 15% and ^{241}Pu to ~ 13% (this one is mainly created by ^{240}Pu conversion). In fact, from the governing equations given in (A.3) through (A.8) and from the σ values given in (A.1) it is easily seen that it is possible to destroy ^{239}Pu, but it is difficult to get a high level of ^{240}Pu cleansing and therefore of ^{241}Pu elimination. If Eqs. (A.6) through (A.8) are solved for a continuous cycle without unloading-reloading, it is seen that the residual fraction of ^{239}Pu follows nearly the law

$$f_q = 10^{-x} \tag{A.11}$$

where $x = \phi t/10^{24}$. However, the residual fractions of ^{240}Pu and ^{241}Pu follows the law

$$f_{0,1} = 10^{-y} \tag{A.12}$$

with y = $\phi t/3\times10^{24}$. Hence, for $\phi t=6\times10^{24}$ (which is a very large value of fluence) the residual fraction of ^{239}Pu would be ~ 1 ppm, while the fraction of ^{240}Pu would be ~ 1.1% and that of ^{241}Pu ~ 0.8%.

Figure A.3 **Accumulated inventory loaded in the transmutator and total amount of transmuted material for the case given in A.1**

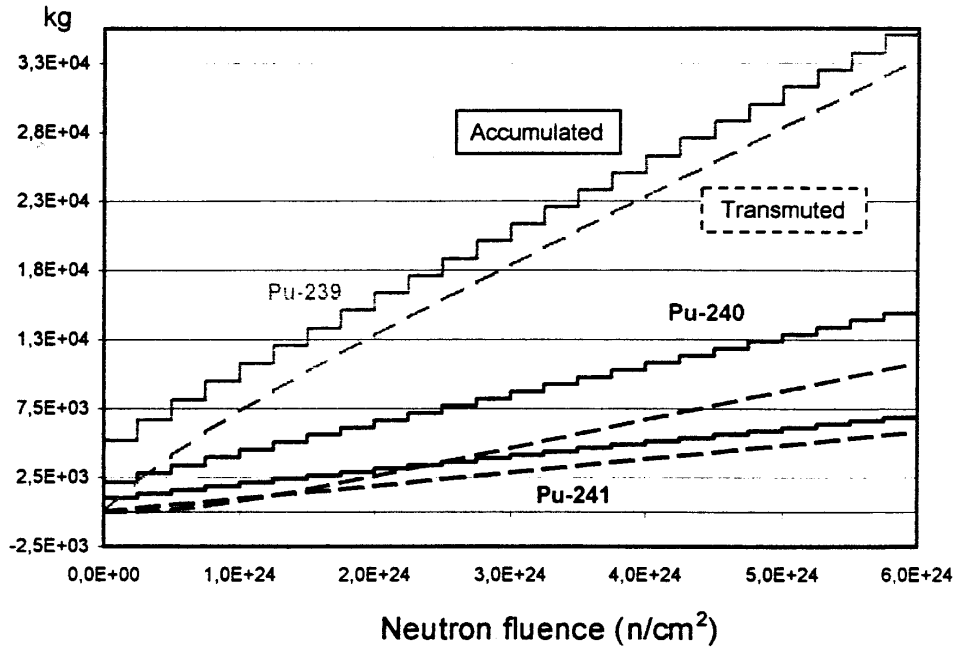
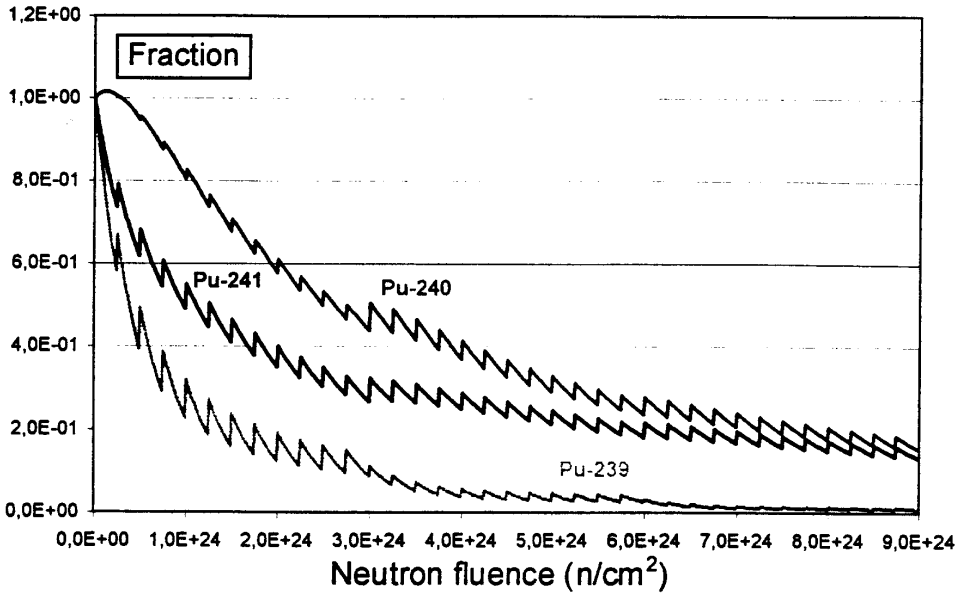


Figure A.4 **Accumulated inventory loaded in the transmutator and total amount of transmuted material for the case of using recycled fuel from the equilibrium cycles of the transmutator after 12 and 24 cycles**



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