AMERICIUM TARGETS IN FAST REACTORS

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Abstract

It would be advantageous to irradiate americium targets up to very large burn-ups, so as to throw them away to waste without any further reprocessing. Former studies considered the recycling of americium in conventional LWRs. But even after long irradiation times the incineration rate of americium, of the order of 50%, was not sufficient to avoid reprocessing and re-fabrication of targets.

In fast reactors, americium targets could be placed in special assemblies filled with a neutron moderator material which enhances Am incineration. Recent calculations show that a burn-up of 90% in the targets could be reached upon affordable irradiation times.

A major problem remains the choice of the inert support material of the targets. So far, the support materials currently envisaged have exhibited too large swelling rates. A type of support material virtually non swelling would be in the form of coated particles, like those irradiated without damage up to very high burn-ups in high temperature reactors. Studies are being launched on the feasibility of such Am targets with coated particles in fast reactors.

1. Introduction

It is a hard challenge in the nuclear fuel cycle to simultaneously insure a sustainable energy supply, and reduce the long-term toxicity of nuclear waste.

The first goal implies to reprocess the spent fuel and to recycle it to obtain the maximum energy generation. This leads to recycle plutonium up to a high burn-up, many successive times. The reactor system has to be (at least) self-sustaining in plutonium.

But the irradiation of plutonium gives rise to a build-up of americium, which in turn produces curium, which will later partly decay to plutonium again. The long-term toxicity of nuclear waste is dominated, for storage times ranging from 300 years to 100 000 years, by Pu, Am and Cm isotopes. In first approximation, the sum of the quantities of Pu, Am and Cm discharged from the reactor, related to the electrical energy produced, may be taken as an indicator of the long-term radio-toxicity.

The reactor type which allows best to extract energy from plutonium is the fast neutron reactor; it also limits to a large extent the Pu isotopic degradation and the formation of Am and Cm. However some minor actinides are still created and should be minimised. In particular, curium is an intense alpha and neutron emitter, so that its presence is a heavy burden in fuel cycle operations (reprocessing, transport, re-fabrication, handling).

In this paper, the following approach is favoured:

- Preference is given to a heterogeneous recycling of minor actinides, separated from plutonium and embedded in special target pins, with a support material inert to neutrons; in comparison, the homogeneous admixing of these actinides in the basic MOX fuel is more cumbersome and penalising.
- It would be an advantage to recycle americium only, leaving curium to the waste, provided however that a long once-through irradiation of these Am targets makes sense, with the aim to avoid if possible any spent target reprocessing.

This paper presents in part 2 the types of fuels and reactors considered. In part 3, different scenarios of energy generation either by PWRs or by LMFRs are considered for recycling, Pu, Am and Cm; a “double-strata” scenario with the introduction of accelerator driven systems (ADS) is also included for comparison.

Part 4 shows how a mixed scenario (PWRs+LMFRs) can be adapted to recycle Pu in the basic fuel, and incinerate Am and Cm, or Am only, in targets. A discussion of all these scenarios is conducted in part 5, with an hint at the penalties incurred in fuel cycle costs and electricity generation costs. Conclusions are drawn in part 6.

The paper makes use of calculations run at Belgonucléaire [1,2] and at CEA [3,4]. The results were sometimes normalised to improve their consistency.

2. Types of fuels and reactors

The paper is centred on oxide fuels, either in thermal or fast reactors.

The reference case for all comparisons is a 1 500-MWe PWR loaded with UO_2, irradiated to an average discharge burn-up of 60 GWd/t, which is the burn-up target for the years to come.

Pu recycling is being practised in PWRs nowadays. But so far, Pu was rarely recycled more than once, because of its important isotopic degradation and production of minor actinides under irradiation with thermal neutrons.

A way to allow Pu multi-recycling in PWR is the so-called MIX concept, in which the MOX Pu content is stabilised by adding enriched UO_2 to the MOX. In this way, Pu multi-recycling can proceed further in PWRs and reach an equilibrium in which Pu consumption equals Pu production. According to calculations, about 10 successive recycles are necessary to reach the equilibrium. Nevertheless, such a solution is costly, as nearly the same UO_2 enrichment is needed as in the reference UO_2 case.

The fast reactor concept considered is the EFR of 1 500 MWe, which was extensively studied by the EFR Associates as a successor to Superphenix [5]. The EFR cores retained, (140 GWd/t average burn-up with the design limit of 200 dpa damage in steel), are either self-sustaining in Pu with a thin fertile blanket, or moderately burning Pu when all fertile blankets are removed. The average Pu enrichment is about 20% in the first case and 23% in the second one.

CAPRA-type EFR cores with very high Pu enrichments of about 40% are not considered here: these strong Pu burners are producing a too large amount of minor actinides.

Table 1 gives the main fuel cycle characteristics and the types of reactors retained. The equilibrium is reached between production and consumption of Pu, or of Pu+Am+Cm.

Table 1. **Fuel characteristics and reactor types (Equilibrium cores)**

Reactor type	**Main fuel enrichment %**	**Fuel mass in reactor** (t/GWe)	**Residence time** (efp days)	**Actinides created (+) or destroyed (-) (kg/TWhe)**		
				Pu	**Am**	**Cm**
PWR (UO_2), 60 GWd/t	4.9 (U5)	86	1 780	+26	+1.65	+0.25
PWR (MIX), 60 GWd/t						
- Pu recycling only	4.5 (U5), 2.1 (Pu)	76	1 560	0	**+4.5**	**+2.24**
- Pu, Am, Cm recycling	4.5 (U5), 2.7 (Pu)	76	1 560	0	**0**	**0**
LMFR: EFR, 140 GWd/t						
- Pu recycling only	20 (Pu)	28.5	1 700	0	+3.5	**+0.28**
- Pu, Am, Cm recycling	20	28.5	1 700	0	0	**0**
- Pu burner	23	28.5	1 700	-20	0	**0**

3. Scenarios of Pu, Am and Cm homogeneous recycling

Calculations were first done with Pu only being recycled in these reactors, while Am and Cm are thrown to waste. Plutonium is supposed to be recovered at reprocessing up to 99.9%, a performance already achieved today. The quantities of Pu, Am and Cm, related to the electricity generation, in kg/TWhe, going to wastes, give an indicator of waste toxicity over storage times varying from 1 000 to 100 000 years.

When recycling Pu but not the minor actinides, PWR and LMFR reactors allow reducing the actinide waste by a factor of 4 or 7, respectively. To reduce them further requires to also recycle minor actinides.

Calculations were thus mainly run for the simultaneous recycling, in the form of a homogeneous MOX fuel, of Pu, Am and Cm. One assumes that, while 99.9% of Pu is recovered at reprocessing, 99% of Am and Cm can be recovered. The respective losses of 0.1 and 1% are coherent targets; as Am + Cm represent less than 10% of Pu in the reference case, it makes sense to assume 1% losses.

The core variants of PWR and FR have slightly larger enrichments than when recycling Pu alone, as they need to be self-sustaining in Pu, Am and Cm.

An interesting mixed scenario is built, combining PWRs, and LMFRs; the exact share results from mass balance calculations. The PWRs fuelled with UO_2 produce the actinides while the LMFRs incinerate them. The LMFR reactor type is an EFR without blankets, with an enrichment of about 23% Pu.

A further scenario is added for the sake of comparison. It follows the double strata principle. The major electricity generation is still provided by a mixed reactor system, PWR / EFR, but the latter recycles Pu only, while special dedicated reactors are assumed to burn minor actinides (they need some plutonium, too). Being fuelled mostly with minor actinides in a dedicated U-free fuel, they will have deteriorated safety coefficients (lower Doppler, larger coolant void reactivity), and this is the reason why they would preferably be accelerator driven systems (ADS), i.e. fast reactors with Keff ~ 0.95, and a neutron spallation source.

Calculations showed that it was possible to minimise the (expensive) second stratum to some 5% of the electricity production.

Table 2 gives the actinide throughputs, in kg/TWhe, for all these scenarios of homogeneous recycling of Pu+Am+Cm.

The first observation is that both all-PWR (MIX) and all-LMFR (EFR) scenarii offer equivalent actinide waste reduction factors, about 130. One noticeable difference is that the all-LMFR strategy recycles more Pu, but 5 to 6 times less Cm.

A mixed scenario with about 34% PWRs fuelled with UO_2 and 66% LMFRs of the EFR type without blankets can give a larger reduction factor, mainly because the recycled Pu quantities are lower. According to the EFR studies [5], the kWhe production cost is about 10% larger for EFR than for a PWR, so that a mixed reactor park is better from the economical point of view.

Surprisingly enough, a double strata strategy with 5% ADS is not better to reduce the actinide wastes. This is explained by the relative accumulation of minor actinides: on multiple recycling the quantities of minor actinides to be loaded remain relatively important. Of course the uncertainty on such quantities at equilibrium is quite large, so that the efficiency could be quoted roughly comparable.

It should be underlined that this conclusion depends on the assumptions made on the recovery rates at reprocessing: 0.1% for Pu and 1% for Am and Cm. If they were taken to be 0.1% for Pu and all Am and Cm isotopes (as many scientists do assume), the comparison would give different results, like:

- A factor 180 for the all-FR scenario.
- A factor 290 for the mixed PWR/EFR scenario.
- A factor 250 for the introduction of 5% ADS in a double strata strategy.

Nevertheless the trends observed above remain the same, with a slight advantage to the mixed scenario.

Table 2.**Homogeneous recycling of Pu, Am, Cm.**
Actinides throughputs (kg/TWhe).

Scenarii	**Core throughputs (kg/TWhe)**					**Actinide waste (kg/TWhe)**				**Waste reduction factors**
	Pu	**Am**	**Cm**	**TRU**	**U+ TRU**	**Pu**	**Am**	**Cm**	**Total**	
100% PWR UO_2	0	0	0	0	0	26	1.65	0.25	27.9	**Ref.**
All-PWR:	56	6.5	8.9	71	2 050	0.056	0.065	0.089	0.210	**130**
All-EFR:	143	5.7	1.6	150	705	0.143	0.057	0.016	0.216	**130**
Mixed PWR(UO_2)-EFR:	98	5.9	2.1	106	463	0.098	0.059	0.021	0.178	**160**
Double strata: Mixed PWR(UO_2)-EFR(Pu) + 5% ADS	99	10.5	3.65	113	381	0.099	0.105	0.036	0.24	**120**

4. Scenarios with minor actinide targets (heterogeneous)

Instead of polluting all MOX fabrication plants with Am and especially Cm, strong emitter of neutrons and of alphas (heating), it is preferable to handle the minor actinides in a distinct fabrication chain with reinforced shielding where minor actinides could be embedded in targets with a support material inert to neutrons.

Further simplifications and cost savings could be obtained:

a) By incinerating the targets in a single long irradiation, up to say, 90% burn-up, and by rejecting the spent targets to waste without reprocessing, so as to avoid the target reprocessing costs (target reprocessing has not been demonstrated so far).

b) By incinerating americium only, and rejecting curium at all stages.

To judge the pros and cons, cost savings are to be put in regard of the actinide waste reduction factors. It is clear that a) and b) lead to an imperfect incineration.

A priori, actinide targets could be inserted in thermal reactors. Calculations were done [2] with the following assumptions about the target pins:

- Loading onto corner positions of MOX assemblies in PWR reactors with the MIX concept (use of enriched ^{235}U to stabilise the Pu enrichment).
- Irradiation time 3 times longer than for the basic fuel pins.

But even with such prolonged irradiations, the results were disappointing, as 50% of the minor actinides loaded had not yet been fissioned at discharge.

For that reason, actinide targets are better loaded in fast reactors; they would be placed in special assemblies of the core, filled with a moderator material (B^{11}_4C, ZrH_2 and CaH_2 have been considered). The presence of moderator improves the efficiency of actinide transmutation. More details on these calculations are given in [4].

Table 3 gives, for the mixed PWR/EFR scenario, the actinide mass balances with the use of targets in EFR, for three different assumptions:

a) Multiple irradiation with intermediate reprocessing of Am+Cm targets, for which a 1% loss will be assumed, in coherence with the homogeneous cases above.

b) One irradiation only of Am+Cm targets up to a 90% burn-up; the spent targets are thrown to waste without reprocessing.

c) One irradiation of Am targets up to a 90% burn-up.

The second case was explicitly calculated [4]. Reaching 90% burn-up was shown to be possible by the insertion of 42 target assemblies in the EFR core which normally contains 388 fissile positions, and by the addition of a complete outer row of 78 target assemblies replacing the radial blanket. The residence time of the 42 inner target assemblies would reach 10 years, to be compared with the 6 years residence time of the EFR fissile assemblies.

Thanks to the introduction of these target assemblies, some important safety parameters are improved, like the Doppler effect (increased) and the coolant void reactivity (decreased); simultaneously, the limit in terms of steel damage (200 dpa) can still be guaranteed.

The results of Table 3 refer to a mixed PWR(UO_2)/EFR scenario: according to the calculations, 44% of the energy would be supplied by PWRs and 56% by FRs of the EFR type without blanket (These are preliminary results, the respective shares of the reactors could still slightly change).

It can be observed that a mixed PWR(UO_2)/EFR scenario with reprocessing gives about the same actinide waste reduction factor (160) in the homogeneous or in the heterogeneous option.

An imperfect target recycling leads to smaller reduction factors of, about respectively:

- 50 if (Am,Cm) targets are incinerated to 90% and not reprocessed anymore.
- 30 if targets loaded with Am only are incinerated to 90% and not reprocessed anymore.

Table 3. **Heterogeneous recycling of Am, Cm in targets. Actinide streams (kg/TWhe).**

Cases	In-core streams (kg/TWhe)					Actinide wastes (kg/TWhe)				Wastes reduction factor
	Pu	Am	Cm	TRU	U + TRU	Pu	Am	Cm[1)]	Total	
Scenario 44% REP UO_2 and 56% EFR[2)]	89			89	394	0.089 +	0.035 +	0.005 +		
Targets: a) Am, Cm reprocessed and recycled		***3.5***	***0.8***			*0.014*	–	*0.029*	0.172	**160**
b) Am + Cm up to 90% burn-up		***3.5***	***0.5***			*0.125*	*0.038*	*0.235*	0.527	**50**
c) Am alone to 90% burn-up		***3.5***	**–**			*0.086*	*0.032*	*0.690*	0.937	**30**

1) In case a, for Cm: 0.465 are rejected from the fuel and 0.230 from the targets.
2) Figures on this line correspond to the core basic fuel; figures of the following lines to the targets.

6. Discussion

The discussion will concern successively: the envisaged scenario, the heterogeneous option, the reduction of waste toxicity, the flexibility of target irradiation and the way to effectively reach 90% burn-up in targets.

6.1 The envisaged scenario

The mixed PWR/FR scenario envisaged above obviously relies on a revival of the fast reactor option. This assumes that fast reactors will be largely deployed, at the time uranium resources will become scarce, and thus more expensive. This was the subject of many studies in the past, and will not be discussed here again.

It will simply be recalled that for the introduction of fast reactors of the EFR type, cost estimates were published by the EFR associates [5]. They essentially set the EFR kWhe cost at a level about 10% higher than for the PWR, and the part of the EFR kWhe cost due to the fuel cycle was estimated to be also about 10%. Such estimates were supposed to apply when EFR type reactors would have largely been deployed.

6.2 Homogeneous or heterogeneous option

Inserting Am or Am + Cm into the MOX fuel itself of the FR in the homogeneous option brings additional difficulties in the re-fabrication plant, so that the fuel cycle cost will increase.

The deliberate insertion of Am in the MOX is an extrapolation from the present fabrication conditions of MOX fuel from an aged Pu. On this effect a comparison of dose rates was made in [6]. But the presence of Cm in the fuel would bring a severe penalty, which can hardly be estimated at present.

The heterogeneous option has the clear advantage to disconnect the fabrication routes of Pu (treated in MOX fuel as presently) and Am + Cm, which would be placed in target rods in a separate smaller fabrication facility. Being smaller, it is easier to shield.

According to the figures of Tables 2 and 3, the streams of minor actinides (in kg/Twhe) are reduced from:

	Am = 5.9	Cm = 2.1	in the homogeneous case
to	Am = 3.5	Cm = 0.8	in the heterogeneous case.

The cumbersome Cm streams have been significantly reduced.

It is clear that two sources of extra-costs can be avoided by, first, renouncing to target reprocessing, and especially by recycling Am only and not Cm. Does it make sense?

6.3 The associated reductions of waste toxicity

Is it useful to reduce the waste toxicity due to actinides by a factor of 30 or 50?

The recent studies on partitioning and transmutation were often setting a waste toxicity reduction factor of 100 as a good target. While the toxicity of the spent LWR fuel comes back to the “natural level”, i.e. that of the uranium ore initially used, after about 200 000 years, a reduction by a factor 100 means that this would be after about 2 000 years. The risk associated with human intrusion is obviously minimised.

A reduction by a factor 30 to 50 means that the doses associated with a human intrusion in the waste storage become comparable to those of a human intrusion into a uranium ore layer, not after 2 000 years, but after some 10 000 years.

Reduction factors by 30 to 50 thus appear to make sense.

On the other hand, such reductions would also be favourable for what concerns the heat release of the waste storage. This aspect deserves further studies.

6.4 The flexibility of target irradiation

The great flexibility of target irradiation in moderated assemblies of the fast reactor can be underlined. Indeed this option could be deployed progressively:

- In a first step, the targets would contain Am only, irradiated in one run, and thrown to waste after discharge: the actinide masses in wastes could be reduced by a factor 30, in a mixed PWR(UO_2)/EFR strategy, for a moderate increase of the kWhe cost only.

- A later step could be to add Cm to the Am targets, and burn them in the same way; the waste reduction factor would somewhat increase to 50, but the cost would also increase.
- Later on, and if the need is recognised, the reduction factor might be improved progressively up to about 160, provided that the targets of minor actinides are reprocessed and recycled.

Such versatility is attractive. The needed R&D programme in support, engaged step by step, remains at a moderate level.

6.5 Ways to reach 90% burn-up in targets

Research has been engaged on Am targets. Many European laboratories have started fabrication and irradiation of such targets. In particular, the EFFTRA-T4 experiment with fabrication made at ITU and irradiation in the HFR reactor at Petten, supported by other partners, has reached an Am burn-up of 28% [7]. A problem was however raised by the large swelling of the spinel matrix, which would not allow much longer irradiation. The EFFTRA partners are searching for improvements.

Among the possible solutions, a promising one is offered by the technology of Pu coated particle fuel, as successfully experienced in the DRAGON experimental reactor [8]: this particle fuel did not swell even after a 60% burn-up corresponding to almost complete depletion of the Pu. The adaptation of this process to Am targets deserves therefore careful feasibility verifications.

Studies have just been started at BELGONUCLEAIRE and EDF to assess the possibility of an adaptation of this type of fuel to Am targets, to make them resistant to the important build-up of gas pressure related to helium and fission products, the goal being to effectively burn 90% of the Am loaded.

7. Conclusions

This paper has underlined how attractive is the concept of putting Am, or Am + Cm targets on special moderated positions of an EFR core. Their irradiation up to 90% burn-up seems feasible in a long but still affordable irradiation.

It was shown by calculations that, if reprocessed and recycled, this target concept in a fast reactor could lead, in a reactor park made of a mix of PWRs (UO_2) and LMFRs, to an actinide waste mass reduction factor of 100 or more.

If the targets can reach a 90% burn-up and are not further reprocessed, this factor decreases to about 50 (for the case of Am + Cm targets) or 30 (targets loaded with Am alone). Such reduction factors, though moderate, already represent sensible improvements of the waste storage conditions.

The flexibility of the concept is an advantage. The research might be first focused on Am target irradiation without reprocessing. It could progressively encompass the addition of Cm, and later the target reprocessing.

A problem remains the integrity of targets with a burn-up as high as 90%. Inert support materials irradiated so far exhibited a large swelling rate. The concept of particle coated fuel, virtually non-swelling, deserves to be examined for application to Am targets. Studies on this concept are starting.

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