The Potential of Pressurized Water Reactors for Recycle of Americium-Curium - 10376

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ABSTRACT

The UK National Nuclear Laboratory, in collaboration with EnergySolutions and other partners, has carried out studies related to americium-curium (Am-Cm) target recycle in a Pressurized Water Reactor PWR). This work, which was carried out in the context of Global Nuclear Energy Partnership (GNEP), was intended to establish whether PWRs could be used to irradiate worthwhile quantities of Am-Cm target rods and what transmutation rates could be achieved. Reactor safety analyses were used to assess the impact on the nuclear design of an equilibrium core containing heterogeneous Am-Cm target rods. The study shows that up to 100 kgHM of Am-Cm can be accommodated in every 18 month refueling cycle without significantly impacting the nuclear design limits. The study demonstrated that a high transmutation rate is achievable for americium, though not for curium. A complementary study examined the broader impact of Am-Cm target irradiation on the PWR fuel cycle and considered how it would fit with some high level strategic goals. This paper reports on the two studies and also discusses how the irradiation of Am-Cm targets might fit within the overall GNEP strategy.

INTRODUCTION

The UK National Nuclear Laboratory (NNL) has been collaborating with EnergySolutions and other partners on a Global Nuclear Energy Partnership (GNEP) study funded by US Department of Energy. Led by EnergySolutions, the collaboration has been studying advanced recycle technologies and how these might be applied in future US commercial reactors. An important element of the advanced recycle technology is the separation of americium and curium (Am-Cm) from spent fuel, which is beneficial in reducing the volume of high level waste in the geological repository. In-reactor irradiation of Am-Cm target assemblies can significantly reduce the mass of americium and the collaboration has looked at the suitability of various different reactor systems for this purpose.

It is envisaged that Am-Cm could be recycled in Pressurized Water Reactors (PWRs) and/or Boiling Water Reactors (BWRs) either in the existing US fleet or in the next generation plants that are expected to be built shortly. Another possibility is the irradiation of Am-Cm in current and projected future CANDU plants in Canada. There is a clear advantage in utilizing existing thermal reactors for this purpose, because these reactors already exist and are potentially deployable for this task in large numbers. While future fast reactors might be better capable of recycling Am-Cm, they will not be available in numbers for a long time yet and the immediate availability of the thermal reactors would be beneficial in ensuring the earliest possible start.

In this context, UK NNL has carried out a feasibility study of Am-Cm burning in a modern PWR. The study looked at the impact of loading up to 100 kgHM of Am-Cm target elements in every 18 month operating cycle on the nuclear design characteristics of the core. The analysis was carried using reactor safety analysis codes and examined the impact on core reactivity, core power distributions, reactivity coefficients, control rod reactivity worths and shutdown margins. The analysis assumed that the Am-Cm target rods would be loaded in unused control rod guide thimble locations. While this would require some

design changes to the fuel assembly, these were not followed up in detail. In the context of a nuclear design study such as this, the precise location of Am-Cm target rods within the assembly is only of secondary interest and a more practicable implementation might be to displace normal UO_2 fuel rods by the Am-Cm rods instead. Provided the ratio of Am-Cm to UO_2 rods is not too greatly different, the nuclear design calculations reported here can be considered largely still valid.

UK NNL also carried out a review of the implications of Am-Cm recycle on the entire fuel cycle and the main conclusions of the study are summarized below. This work also identified some high level strategic goals that any transuranic recycle scheme should preferably be able to achieve. A preferred operating domain was identified into which the PWR Am-Cm recycle scheme actually falls. Finally, the study discusses how Am-Cm recycle in PWRs might fit within the overall GNEP strategy in a complementary role with Am-Cm irradiation in CANDU plants. This work therefore assesses the potential role of modern LWRs to manage the historic and future waste arisings; a diverse and potentially pragmatic partial solution to the waste problem.

AM-CM RECYCLE IN PRESSURIZED WATER REACTOR

This section summarises the reactor safety and performance analysis of a 4-loop PWR in which 100 kg of Am-Cm target rods are loaded in every 18 month refueling cycle.

Description of PWR Core and Am-Cm Target Loading Strategy

The PWR core chosen for this analysis was a standard Toshiba/Westinghouse 1200 MWe 4-loop plant; this plant is representative of the existing PWR fleet in the USA and is not dissimilar to potential new build plants and is therefore an appropriate example to use. The objective is to demonstrate in a generic fashion what is achievable in PWR cores in the way of Am-Cm recycle and transmutation and how the presence of the Am-Cm target rods affects the core nuclear design parameters.

The starting point for the analysis assumes that the core is fuelled entirely with UO₂ assemblies with a mean initial enrichment of approximately 4.2 w/o. With an 18 month refueling interval the equilibrium cycle requires 84 feed assemblies to achieve a cycle burnup of 18.3 GWd/t, as calculated with the Core Management System reactor analysis package from Studsvik Scandpower [1, 2, 3]. The equilibrium discharge burnup is approximately 42 GWd/t. The reference loading pattern of the core is designed such that all the nuclear design parameters fall within their respective Nuclear Design Limits. This reference loading pattern is then used to determine the impact of Am-Cm target rod loading without any further loading pattern optimization.

A strategy was now needed for loading Am-Cm target rods in the core. The approach adopted for this study was to load the Am-Cm target rods inside the 24 control rod guide tubes of fuel assemblies not in an Reactivity Control Cluster Assemblies (RCCA) location. The Am-Cm target rods chosen for the study contain AmO_2/CmO dispersed in a ceramic inert matrix. The inert matrix pellets were taken to have an outer diameter of 0.8192 cm. The Am-Cm target rods were assumed to be clad in Zircaloy-4 to give an overall diameter of 0.968 cm, which just fits within the RCCA guide tube inner diameter of 1.1228 cm. It was recognised that this approach is not likely be practical because the Am-Cm rods will be heat generating and there are will be difficulties ensuring sufficient coolant flow to the Am-Cm rods within the tight RCCA guide tubes. A more practical alternative would be to displace a similar number of normal UO_2 rods with Am-Cm rods and to increase the initial enrichment of the UO_2 rods to compensate. However, developing a practical design was outside the scope of work and it was judged that the two approaches are not likely to be too dissimilar neutronically and for the purposes of this study the guide tube loading strategy was preferred.

The isotopic composition assumed for the Am-Cm corresponds to that discharged from a PWR fuel assembly irradiated to 50 GWd/t and cooled for 6 years. Americium accounts for 89.6 w/o of the mix (62.4 w/o Am-241; 0.4 w/o Am-242 and 26.8 w/o Am-243), while Cm-244 accounts for most of the remaining mass. The Am-Cm rod design has a mass of 175.2 gHM per target pin and 4.2 kgHM per assembly. With the 84 assembly refueling strategy, it is possible to accommodate up to 24 assemblies per reload which equates to an Am-Cm loading of approximately 100 kgHM in every 18 month cycle.

Am-Cm Target Behavior

In some respects the behavior of the Am-Cm target rods can be likened to burnable poison rods. Initially they will depress the reactivity of an assembly and the reactivity hold-down will decrease as the Am-Cm is depleted under irradiation. One of the design aims was identified was to obtain an initial reactivity hold-down comparable with existing burnable absorber designs, because it is known that this is compatible with producing a satisfactory core design. Figure 1 compares the infinite multiplication factor versus burnup profile for the assembly containing 24 Am-Cm rods with those for an unpoisoned 4.2 w/o UO_2 assembly and for a similar assembly containing 24 Wet Annular Burnable Absorber (WABA) rods.

As can be seen from Figure 1, the initial reactivity hold-down in the assembly containing Am-Cm rods closely matches that of the assembly containing WABA rods. However, the subsequent burn-out of is very slow for the Am-Cm assembly and this implies there will be a significant residual absorption penalty. This is unavoidable with Am-Cm and would demand that the utility either accepts a shorter cycle length for a given initial enrichment or specifies a higher initial enrichment. Interestingly, the Am-Cm assembly has an elevated k-infinity at the highest burnups, which is indicative of plutonium production in the Am-Cm target rods. This offsets the residual absorption penalty, but is insufficient to completely annul it.

The burn-out characteristics of the Am-Cm rods is of particular interest in this study. Figure 2 shows how the various americium and curium isotopes deplete as a function of the mean burnup of the assembly containing the Am-Cm rods. The intent, of course, is that americium and curium should deplete as completely as possible. Am-241 and Am-243 do indeed display the desired behaviour, with Am-241 in particular almost completely transmuted by 50 GWd/t. However, in common with other studies in both thermal and fast spectrum systems, the curium isotopes accumulate with burnup.

The reactor analyses show that by the time the fuel assemblies containing Am-Cm are discharged from the core, there is net transmutation of approximately 70% of the Am-Cm nuclides. Therefore in every 18 month fuel cycle there is net transmutation of 70 kg of the 100 kg of Am-Cm loaded. As discussed later, this is a quite favorable transmutation rate. Since the equilibrium discharge inventory of Am-Cm from the UO_2 fuel assemblies in a 1.2 GWe PWR is approximately 50 kgHM, this implies that the 100 kgHM Am-Cm load would be sufficient to recycle the Am-Cm arisings from two 1.2 GWe PWRs.



Figure 1: k-infinity versus burnup curves for unpoisoned UO₂ assembly, UO₂ assembly with 24 Wet Annular Burnable Absorber (WABA) rods and UO₂ assembly with 24 Am-Cm rods



Figure 2: Burnup evolution of americium and curium isotopes in Am-Cm target rods.

Impact on Core Design

This section summarises the main results of the reactor safety analysis for the equilibrium core containing Am-Cm target rods. The equilibrium cycle length is reduced by 1.2 GWd/t relative to the reference core design (17.1 GWd/t compared with 18.3 GWd/t). This is caused by the residual absorption penalty of the Am-Cm rods noted earlier. The shortfall in cycle length could either be accepted by the utility, thereby incurring an operational cost penalty, or alternatively the initial enrichment of the UO₂ fuel could be increased slightly (thereby incurring a modest, but still significant fuel cost penalty).

An extensive survey of the nuclear design parameters of the Am-Cm core was carried out, as would be required as part of the normal licensing process. Table I summarises qualitatively the impact that loading Am-Cm target rods has on the nuclear design limits of the core relative to the reference core.

Parameter	Am-Cm impact	Significance
Radial power peaking factor $F_{\Delta H}$	Small	Probable mitigation with core
		loading pattern optimization
		and/or relaxation of limits
Axial power peaking factor F _Z	Small	None
Overall power peaking factor F _Q	Small	Probable mitigation with core
		loading pattern optimization
		and/or relaxation of limits
Moderator temperature coefficients	Marginal violation of Nuclear	Probable mitigation with core
	Design Limit	loading pattern optimization
	G 11 ·	and/or relaxation of limits
Doppler coefficient	Small increase	None
Boron coefficients	Slight reduction in magnitude	Probable mitigation with core
	(less negative), marginal violation	loading pattern optimization
	of Nuclear Design Limits	and/or relaxation of limits
Control rod reactivity worths	Slight increase in magnitude	None
	(more negative) within Nuclear	
	Design Limits	Nama
Shutdown margins	Small increase in end-of-cycle	None
Delessed a sector of free stien	snutdown margin	Nama
Delayed neutron fraction	Small	None
Boration limits	Moderate increases in boron	INONE
	concentrations within Nuclear	
	Design Limits	

Table I : Summary table of impact of Am-Cm target loading on nuclear design parameters

As can be seen, the radial and axial power distributions are perturbed relative to the reference core, but only to a small extent. Four of the nuclear design parameters slightly exceed the Nuclear Design Limits, but the violations are sufficiently small that they could easily be mitigated with minor optimization of the core loading pattern. These results give confidence that loading up to 100 kgHM of Am-Cm targets per 18 month fuel cycle in a PWR would be feasible.

FUEL CYCLE AND STRATEGIC ISSUES

While the nuclear design study has demonstrated initial feasibility of Am-Cm target irradiation in PWRs, it is important not to lose sight of the broader implications for the PWR fuel cycle. A complementary

study was carried out which reviewed the impact of Am-Cm recycle on all aspects of the fuel cycle. The study also carried out technology readiness assessment for each stage. Finally, the strategic fit of Am-Cm recycle was assessed against some high level strategic criteria that were derived for that purpose.

Fuel Cycle Impact and Technology Readiness

The starting point for the fuel cycle survey was to assume that a conventional nuclear fuel recycle scheme is already in place and the purpose is to identify what aspects of this fuel cycle are impacted by Am-Cm burning. Irradiated PWR fuel is discharged into the cooling ponds, transported to the recycle plant (after allowing sufficient cooling time prior to transport) and then stored in the fuel receipt ponds prior to fuel shearing operations. Subsequent to fuel shearing the recycle process is unaffected up to the point at which Am-Cm is separated from the uranium plutonium and high level waste streams. This is the point at which the Am-Cm fuel cycle diverges from the normal U-Pu recycle schemes currently implemented commercially in France, Russia, Japan and UK.

The review identified ten key stages of the fuel cycle which will be significantly impacted by Am-Cm recycle:

- 1. Recycle plant design and operations
- 2. Storage and on-site transport
- 3. Target fabrication
- 4. Recovery of residues and out-of-specification material
- 5. Transport and receipt at reactor site
- 6. Target loading
- 7. Reactor operations
- 8. Irradiated target handling/storage
- 9. Irradiated target conditioning/disposal or recycling
- 10. Waste management

For each of these stages a technology readiness assessment identified that significant or major technological development would be required for all ten areas and that for none of these areas is the required technology already established. Under several of these headings, recurring technical themes requiring major development were those of logistics and safety/licensing. A recurring non-technical theme was that of stakeholder engagement, because many of these areas will require the construction and operation of large facilities that will represent a significant departure from current practice in the nuclear industry and which will therefore demand very carefully argued justification.

Strategic fit

This section considers how Am-Cm target recycle in PWRs fits within the overall fuel cycle and assesses how well suited PWRs are to this role. For this purpose a simple parametric analysis is very useful based on two simple parameters:

- 1. The burnup goal Ω , defined as the targeted fraction of minor actinides to be destroyed. Achieving complete destruction $\Omega = 1$ or near complete $\Omega = 0.99$ are probably impractical and more realistic targets might be in the range $\Omega = 0.80$ to 0.95. The final target might need to be established as a compromise between what is desirable and what is practicable.
- 2. The depth of burn ε , defined as the fraction of minor actinides destroyed in a single target irradiation lifetime. This is a performance measure of the specific target design and the reactor system in which the targets are irradiated. For the particular PWR Am-Cm target designs PWR,

after three irradiation cycles 70 kg of Am-Cm is destroyed by the time the targets are discharged for every 100 kg initially loaded. Therefore for this system $\varepsilon = 0.7$ and if the burnup goal Ω exceeds this value, then a multiple recycle strategy is necessary.

It is desirable that the depth of burn ε parameter should be as high as possible to minimise the number of recycles required to meet the burnup goal Ω . The number of recycles n required is given by:

$$(1-\varepsilon)^n = 1-\Omega$$
$$n = \ln(1-\Omega)/\ln(1-\varepsilon)$$

Figure 3 shows a plot of n (y-axis) versus ε (x-axis) for $\Omega = 0.8$, 0.9, 0.95 and 0.99. The number of recycles becomes impractically large for small ε (< 0.5) and high values of Ω . For n = 5, the elapsed time to burn an initial sample down to the burnup target approaches 50 years or so and for n > 5 the entire scheme can be considered impractical. This is the reason for the preferred domain indicated at the right of the graph. The PWR with $\varepsilon \sim 0.7$ actually falls well inside the preferred domain and even an ambitious burnup target of $\Omega = 0.95$ is achievable with only about three recycles, meaning that the target can be met within a reasonable elapsed time.

It is also desirable to minimise the total inventory of Am-Cm that is actively under irradiation. The whole justification for irradiating Am-Cm is that it is considered to be one of the most hazardous by-products of fission reactors. Although loading this hazardous material in a reactor core introduces a short term risk, the argument is that this is balanced by the long term benefits of Am-Cm destruction. Nevertheless, good practice requires the minimization of the hazard potential presented by in-reactor irradiation and this implies minimizing the in-core Am-Cm inventory.

For a starting mass M (kg) the Am-Cm inventory in a multiple recycle scheme (with n recycles) at the start of the equilibrium cycle the End of Cycle (EOC) mass is given by:

EOC Mass = M{
$$(1-\epsilon) + (1-\epsilon)^2 + ... + (1-\epsilon)^n$$
} = M[1-(1- $\epsilon)^n$][1- ϵ]/ ϵ



Figure 3: Number of recycles (n) required to achieve burnup target ranging from $\Omega = 0.8$ to $\Omega = 0.99$, as a function of ε

Figure 4 shows a plot of the EOC mass factor as a function of depth of burn ε for the same target burnups $\Omega = 0.8, 0.9, 0.95$ and 0.99. The preferred domain is on the right of the graph again, where the active EOC mass is less than the mass originally loaded. To the left of the preferred domain, the EOC mass factor in the equilibrium cycle is > 1, so that for every 1 kg of Am-Cm loaded in the first recycle there remains more than 1 kg in active irradiation in the core. With $\varepsilon = 0.3$, the active mass is double the original mass and for very small ε the active mass climbs steeply. The PWR with $\varepsilon = 0.7$, is very firmly in the preferred domain and the active mass factor remains less than 1.0 even for very high values of Ω .

From both Figure 3 and Figure 4 the clear conclusion is that with a depth of burn of 0.7, PWR ranks very favorably for Am-Cm burning in that a substantial proportion of the Am-Cm loaded is transmuted within a single target irradiation lifetime and that it fits within the preferred domains.

Another reason for preferring a small number of recycles is that every recycle incurs a cost. If the cost is expressed in terms of Dollars per kgHM, then the cost incurred at each recycle is proportional to the Beginning of Cycle (BOC) mass plotted in Figure 5 that is given by:

BOC Mass = M{1+
$$(1-\varepsilon)$$
 + $(1-\varepsilon)^{2}$ +...+ $(1-\varepsilon)^{(n-1)}$ } = M[1- $(1-\varepsilon)^{n}$]/ ε

It is desirable that the ε parameter should not be less than 0.5, which is the region below which the BOC mass starts to escalate rapidly.



Figure 4: End of Cycle (EOC) active mass in kg for every 1 kg loaded for burning target ranging from $\Omega = 0.8$ to $\Omega = 0.99$, as a function of ϵ



Figure 5: Beginning of Cycle (BOC) active mass in kg for every 1 kg loaded for burning target ranging from $\Omega = 0.8$ to $\Omega = 0.99$, as a function of ε

POTENTIAL ROLE OF AM-CM BURNING IN THERMAL REACTORS

The major isotopes of americium and curium are heat generators that potentially limit the utilization of repository capacity for disposal of HLW. Wigeland et al [4] has indicated that repository capacity utilization could be improved by at least a factor of five upon separation of plutonium and americium from spent nuclear fuel. Transmutation of the separated americium has traditionally been considered only in fast reactors, which are several decades from commercialization. Therefore, americium transmutation in currently deployed thermal reactors represents a nearer term opportunity for managing this actinide and a viable alternative to fast reactor transmutation. Note that the significant heat generating curium isotopes have relatively short half-lives and their destruction is probably better accomplished by natural radioactive decay. However, the separations technology deployable in the near term cannot separate americium from curium and so the two actinides exist as a mixture.

The core design study discussed in Section 2 has shown that there is the potential to recycle up to 100 kgHM of Am-Cm per 18 month cycle and that by the time the Am-Cm target rods have achieved their full discharge burnup 70% of the Am-Cm will have been transmuted. As part of the GNEP collaboration, Atomic Energy of Canada Ltd (AECL) carried out a similar study for a CANDU-6 reactor. This study indicated that CANDU-6 might be capable of achieving rather higher Am-Cm loadings, despite the smaller electrical capacity of the CANDU plant. A range of different Am-Cm recycle options were considered by AECL, including homogenous and heterogeneous loading options and with different Am-Cm concentrations in the target bundles. A range of Am-Cm loadings up to 875 kg/year were indicated, though the depth-of-burn values achievable were all lower that the 70% indicated for PWR. In fact, there is a trade-off between maximizing Am-Cm loading and depth of burn with the maximum Am-Cm loading being achieved at the expense of depth of burn. The higher Am-Cm loadings in CANDU-6 are possible because more core locations are potentially available than the vacant control rod guide thimbles in the PWR study (for the homogenous core loading potentially all the fuel channels are available, while for the heterogeneous core loading one in every four fuel channels are available). The smaller depth-of-burn values in CANDU results from the lower discharge burnups achievable in the Am-Cm target rods in CANDU-6. Recycling the irradiated target rods to recover residual Am-Cm for re-loading into new rods is being considered to optimize overall actinide transmutation.

The PWR and CANDU studies both indicate that worthwhile Am-Cm loading and transmutation rates can be achieved in thermal reactors. It is estimated that the total Am-Cm inventory in LWR spent fuel discharged in the USA up to 2001 will be 53 tHM by the earliest time US recycle plants could be operational in 2030. This entire inventory could be recycled in program of approximately 750 reactor-years of operation of a 1.2 GWe PWR, which could be achieved for example with 25 PWRs implementing Am-Cm recycle for 30 years each. This would be a difficult logistical challenge, but is nevertheless technically feasible. Alternatively, the same recycle rate could be achieved with about 60 reactor-years of CANDU-6 operation, though with a much lower depth-of-burn (higher depth-of-burn values could be achieved in CANDU-6 at the expense of lower Am-Cm loadings and therefore more reactor-years). Regulations restricting management of foreign radioactive material in Canada could be overcome by deploying a CANDU-6 reactor dedicated to transmuting Am-Cm in the USA. In either case, the limiting factor may not be the reactors themselves but rather the availability of Am-Cm from the recycle plants and the fabrication capacity for Am-Cm target rods.

While it is possible that future advanced reactors might be able to out-perform PWR or CANDU at Am-Cm recycle, these thermal reactors have an outstanding advantage in that they are already in operation. From a technical standpoint, they are potentially deployable in this role in the near term, with no need for the extensive technical development and commercialization characteristic of advanced reactors. Americium transmutation in LWRs and CANDU reactors is technically feasible at a rate sufficient to destroy the americium present in the existing used nuclear fuel stockpile and future arisings. Both reactor types fulfill complementary and mutually supporting roles in this function and their respective use will be driven by commercial, regulatory and political considerations.

CONCLUSIONS

- A 1.2 GWe PWR has been shown capable of recycling 100 kgHM of Am-Cm every 18 month fuel cycle. Am-Cm target rods were assumed to be loaded in the RCCA guide tubes of a conventional PWR fuel assembly. This is acknowledged as possibly not being technically feasible, but is considered a sufficiently good representation to indicate what the core is potentially capable of in a more practical design (such as displacing a fraction of the conventional UO₂ rods with Am-Cm target rods).
- After the full irradiation of the Am-Cm targets is complete, the analysis indicates that the depth-ofburn parameter ε is in the region of 0.7. The equilibrium core mostly satisfies the nuclear design limits and though four were exceeded, the violations were quite marginal and probably can be mitigated by minor adjustments and loading pattern optimization.
- The Am-Cm target behavior can best be considered analogous to that of a burnable absorber, though with a significant residual absorption penalty. The effect of loading 100 kgHM of Am-Cm per 18 month cycle without a compensating increase in the enrichment of the UO₂ fuel rods is a loss of cycle length from 18.3 GWd/t to 17.1 GWd/t, which is significant and represents a direct loss or cost to the utility.
- While the americium in the Am-Cm targets displays good transmutation behaviour, the curium component accumulates rather than being burned. This is consistent with other studies of curium transmutation and does not just result from the thermal spectrum.
- A review of ten key stages of the PWR fuel cycle impacted by Am-Cm recycle highlighted that significant or major technological development was required in all ten areas and that for none of these areas is the required technology already demonstrated.
- Some high level strategic targets were derived for Am-Cm recycle and the PWR Am-Cm recycle option was found to fall inside the preferred operating domains and can therefore be regarded as very favorable.
- A discussion of the potential role of Am-Cm recycle in PWR highlighted that it is technically feasible to recycle Am-Cm from historic stocks of spent fuel in the USA and from future spent fuel arisings as well. Comparison with a similar evaluation carried out for CANDU-6 highlighted a degree of overlap between the two systems, with CANDU showing potentially greater capability for large Am-Cm loadings. The potential for PWR and CANDU plants to be deployed in a complementary role, which is necessary because the CANDU plants on their own are unlikely to have the capacity required to recycle all the historic and future Am-Cm arisings from the USA.
- The transmutation of Am-Cm in LWRs is a potential alternative to direct disposal as a waste form.

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