REDUCING THE IMPACT OF USED FUEL BY TRANSMUTING ACTINIDES IN A CANDU REACTOR

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Abstract

With world stockpiles of used nuclear fuel increasing, the need to address the long term utilization of this resource is being studied. Many of the transuranic (TRU) actinides in nuclear spent fuel produce decay heat for long durations, resulting in significant nuclear waste management challenges. These actinides can be transmuted to shorter-lived isotopes in CANDU® reactors to reduce the decay heat period.

Many of the design features of the CANDU reactor make it uniquely adaptable to actinide transmutation. The small, simple fuel bundle facilitates the fabrication and handling of active fuels. Online refueling allows precise management of core reactivity and separate insertion of the actinides and fuel bundles into the core. The high neutron economy of the CANDU reactor results in high TRU destruction to fissile-loading ratio.

This paper provides a summary of actinide transmutation in CANDU reactors, including both recent and past activities[1]-[4]. The transmutation schemes that are presented reflect several different partitioning schemes and include both homogeneous scenarios in which actinides are uniformly distributed in all fuel bundles in the reactor, as well as heterogeneous scenarios in which dedicated channels in the reactor are loaded with actinide targets and the rest of the reactor is loaded with fuel.

1. Introduction

Management of spent nuclear fuel will continue to be a challenge in the future, as stockpiles increase worldwide. Many of the transuranic (TRU) nuclides (Pu, Am, Cm, Np) are long-lived, and continue to produce heat for thousands of years. This long-term decay heat presents a challenge to the disposal of spent nuclear fuel. Reducing the decay heat of spent nuclear fuel (SNF) from light water reactors (LWR) will increase the capacity of long-term geological disposal sites.

The CANDU reactor offers attractive solutions for effectively dealing with used nuclear fuel from a LWR fleet. Many of the design features of the CANDU reactor make it uniquely adaptable to actinide transmutation as well as utilization of LWR used fuel with minimal reprocessing. The most significant feature is the high neutron economy resulting from the heavy water moderator, which allows a high TRU destruction rate relative to the fissile loading because more neutrons are available for transmutation rather than being parasitically absorbed in the moderator. Another important feature of the CANDU reactor is that the refueling is performed on-power and separately for each fuel channel. This allows actinide targets to only occupy desired locations in the reactor and the residency time of the targets to be adjusted separately

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from regular fuel bundles. Online refueling also allows precise management of core reactivity, and further increases the neutron economy relative to batch refueling.

Lastly, the small and simple CANDU fuel bundles, short in length and light in weight, facilitate the fabrication and handling of active fuels. These characteristics also enable a CANDU fuel bundle to function as a target carrier with minimal or no design change to the bundle.

The introduction of minor actinides into the reactor for transmutation could be achieved in many different configurations. These are generally grouped into three main categories: heterogeneous bundle designs, homogeneous bundle designs, and heterogeneous channel designs. Heterogeneous bundle designs contain two (or more) different fuel types within the bundle. Typically the minor actinides (MA) (Am, Cm, Np) are loaded in the centre pin, or the centre pin and the inner ring, of the bundle. Placing the MA in the centre of the bundle has the additional benefit of reducing the coolant void reactivity. Homogeneous bundle designs contain TRU throughout the entire bundle. The heterogeneous and homogenous bundle designs would be fuelled into the reactor such that every channel is filled with that fuel bundle design.

The fuel channel feature of the CANDU reactor allows for an additional flexibility, in that it is possible to load different fuels in different channels within the reactor, and to have different residence times for the different fuels. The fuelling scheme can be optimized, making use of the change in neutron flux levels in different regions of the core. For example, minor-actinide bearing fuels could be placed in the low-flux outer region of the core where they would have little impact on the remainder of the reactor, and could remain there for much longer than the normal residence time to achieve high levels of transmutation of the MA.

In addition to the many ways to introduce TRU to the reactor, many different reprocessing schemes can be employed to obtain any particular combination of transuranics. Each element could be used separately, or they could be extracted together. Several different partitioning schemes are used in this paper, which correspond to varying levels of complication in reprocessing. In general, schemes which separate out each element individually require more steps, are much more difficult, expensive, and produce more wastes than schemes which keep several elements together in one reprocessing stream.

This paper provides a review of actinide transmutation schemes that have been studied previously [1], [2], [3], [4]. The methods discussed in this paper are listed below:

- Am, Cm, Np, and lanthanides (Ln) loaded in the centre pin of fuel bundle,
- Americium mixed with low enriched uranium (LEU) loaded in the full core,
- Group-extracted TRU (Pu, Am, Np, Cm) in homogenous bundles, and
- 14~25 wt% Am/Cm in an inert matrix placed in 30 periphery channels while the core is fuelled with recycled uranium (RU) from reprocessed used LWR fuel at 0.9 wt% enrichment.

It should be noted that all scenarios presented in this paper are at the research stage and much further development and engineering work would be required to bring these concepts into reality.

1.1 Actinide Transmutation Pathways

One of the key strategies to deal with a long-lived actinide, for example Am-241, is to transmute it (see Figure 1). The transmutation causes decay heat generation to occur in a relatively shorter period after the used fuel is discharged from the reactor. There are several different ways of introducing Am/Cm into a CANDU reactor for transmutation. As shown in Figure 1, the transmutation of Am-241 follows several pathways that affect the decay heat production of the spent fuel, and result in the production of isotopes of curium and plutonium. In the first step, a neutron captures onto Am-241, creating Am-242 or Am-242m.

Several different pathways are available after the initial neutron capture. Am-242m has a high fission cross-section, so by this path the Am can be transmuted by fission. In the second pathway the Am-242 beta decays into Cm-242. The Cm-242 then alpha decays with a relatively short half-life (163 days) and some of the original americium will end up as Pu-238. The Am-242m can also neutron capture to Am-243, and a second neutron capture creates Am-244 or Am-244m. The Am-244 nuclides both have short half-lives and beta decay to Cm-244. Cm-244 has a relatively short half-life, and alpha decays to Pu-240. Am-242m can also decay by electron capture to Pu-242. The isotopes Cm-242, Cm-244 and Pu-238 all have an impact on the decay heat of the spent fuel.

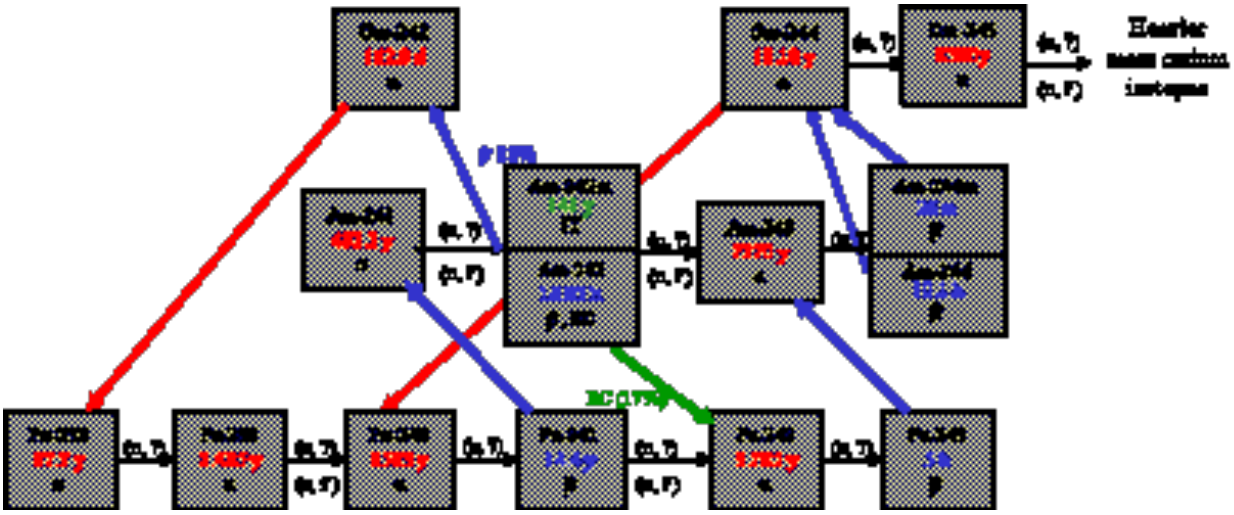

Figure 1. The main transmutation pathways of transuranic nuclides

Am-242m, Cm-245, Pu-239, and Pu-241 are the fissile isotopes. The other isotopes act as a poison, capturing neutrons and reducing the coolant void reactivity (CVR) of the bundle (if located in the centre of the fuel bundle). This process breeds plutonium through the path shown in Figure 1.

With regard to curium production, for schemes with relatively short irradiation time (a few years) the curium nuclides that are created are the low-mass, short-lived curium isotopes, Cm-242 and Cm-244. These isotopes have short half-lives on the same time scale as fission products, and once produced in the used fuel the curium could be stored and decayed similarly to fission products, rather than put into long-term storage or further transmuted.

The destruction of minor actinides (Pu, Np, Am, Cm) is achieved through fission, resulting in fewer minor actinides requiring long-term waste disposal. The calculations presented here were carried out using WIMS-AECL [5] with a nuclear data library [6] with Z up to 96.

2. Heterogeneous Bundle Containing Americium, Curium, Neptunium, and Lanthanides

The transmutation scheme studied in this paper places an americium/curium/neptunium/ lanthanide (elements with Z= 57 to 71) mixture into the centre pin of a CANFLEX® [7] fuel bundle, Figure 2. This would greatly reduce the complexity of the partitioning scheme and potentially significantly reduce the cost of reprocessing. The amount of the actinide/lanthanide (An/Ln) mixture in the centre pin was varied by changing the packing density to 5%, 25%, 50%, 75% and 100% of a nominal density of 10 g/cc. This centre pin can be removed and reinserted into a new fuel bundle for further irradiation to achieve higher total transmutation of the actinides. The rest of the fuel bundle was comprised of 1.0% LEU. This bundle design allows for the transmutation of americium and curium while reducing the coolant void reactivity. The isotopic composition of the An/Ln mixture is given in Table 1. This used nuclear fuel is from a light water reactor (LWR), which was cooled for 10 years before reprocessing. The LWR had an initial enrichment of 4 wt% U-235 and an exit burnup of 50 MWd/kg initial heavy elements (IHE). Only nuclides of interest to reactor physics, that is those with significant neutron cross-sections, are contained in the WIMS-AECL library that was used for the study; other lanthanides present in the spent fuel have been ignored in this work.

Two different cooling times between irradiations are investigated, 3 months and 2 years. This scheme provides the dual benefit of transmuting long-lived actinides into shorter-lived fission products, while at the same time reducing the void reactivity coefficient of the reactor.

The transmutation rate (kg/year) and the cumulative percentage of americium that is transmuted for each recycle of the centre pin are shown in Figure 3 for each density for the case with 2 years of decay between irradiations.

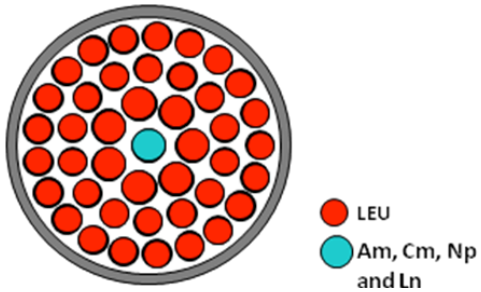

Figure 2. Design for CANFLEX fuel bundle with centre target

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Table 1
Isotopic composition of the Am, Np, Cm, and Ln in the centre pin

Nuclide	% by weight	Nuclide	% by weight	Nuclide	% by weight
Np-237	4.2216	Nd-145	5.7903	Gd-152	0.0005
Am-241	4.5298	Nd-146	6.5062	Gd-154	0.1638
Am-242m	0.0074	Nd-148	3.3197	Gd-155	0.0417
Am-243	1.6005	Nd-150	1.6472	Gd-156	0.9918
Cm-243	0.0038	Pm-147	0.0970	Gd-157	0.0013
Cm-244	0.4893	Sm-147	1.8937	Gd-158	0.2124
Cm-245	0.0488	Sm-148	1.2773	Gd-160	0.0107
Cm-246	0.0062	Sm-149	0.0261	Tb-159	0.0246
Cm-247	0.0001	Sm-150	2.9653	Dy-160	0.0035
La-139	10.9317	Sm-151	0.1010	Dy-161	0.0034
Ce-140	11.5133	Sm-152	0.8101	Dy-162	0.0026
Ce-142	10.0322	Sm-154	0.3483	Dy-163	0.0020
Ce-144	0.0004	Eu-151	0.0082	Dy-164	0.0005
Pr-141	10.0022	Eu-152	0.0002	Ho-165	0.0009
Nd-142	0.2396	Eu-153	1.1507	Er-166	0.0003
Nd-143	6.5302	Eu-154	0.1126		
Nd-144	12.3169	Eu-155	0.0121		

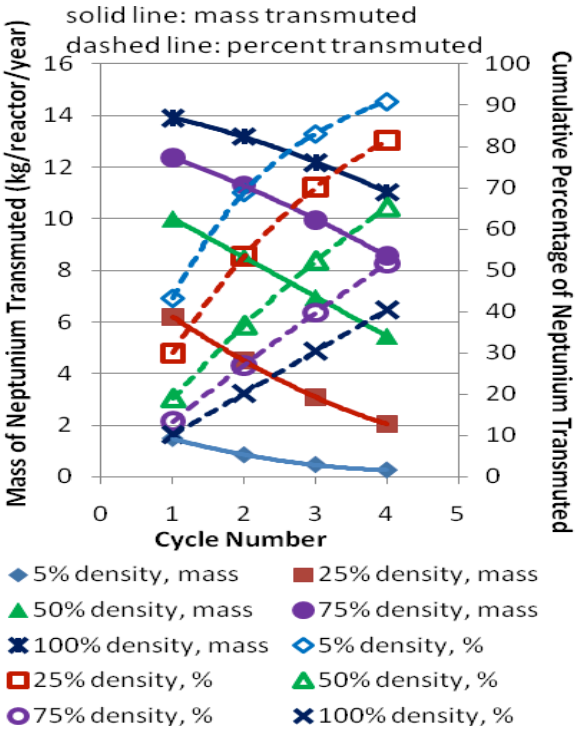

Figure 4. Rate of transmutation of Np

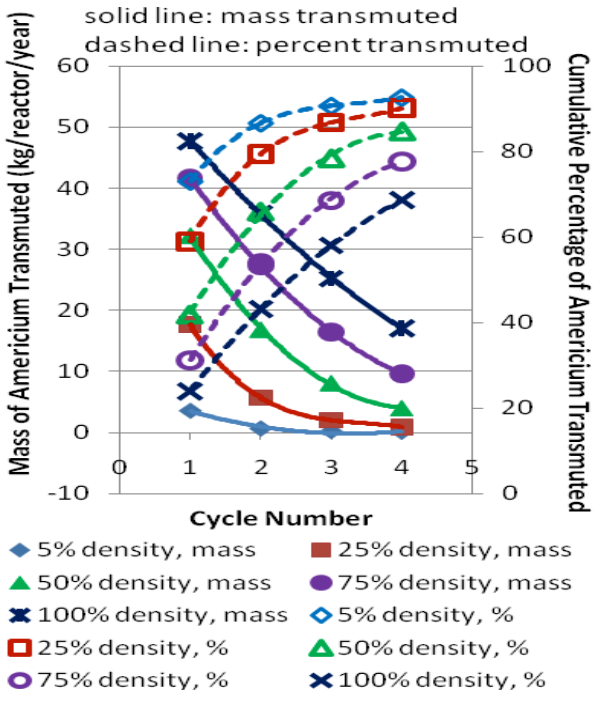

Figure 3 Rate of transmutation of Am

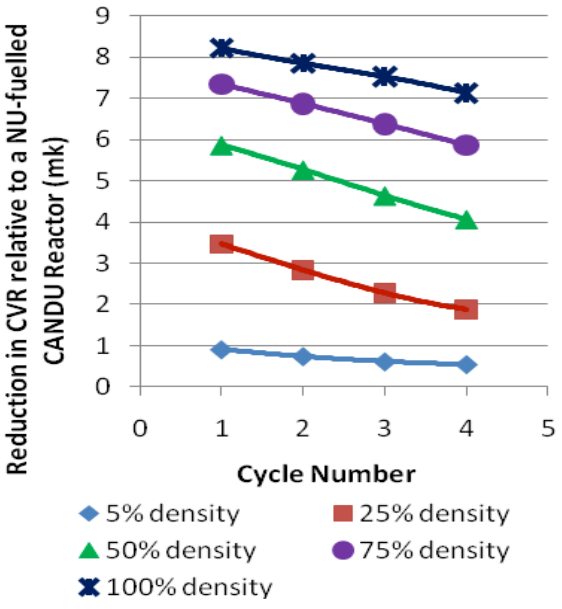

Figure 5. Reduction in CVR with respect to an NU-fuelled CANDU reactor

The decay time between irradiations has little effect on the americium transmutation, less than 1%. As is typical with actinide transmutation scenarios, there is a trade-off between the percentage of the initial americium that can be transmuted and the total mass of americium that can be transmuted. That is, if a higher initial mass is loaded in the reactor, then more total mass will be transmuted, but a larger fraction will remain at the end of the irradiation than if a smaller initial mass is used. For the 5% and 100% cases respectively 93% and 69% of the initial americium is transmuted at average rates of 1.1 and 31.5 kg/reactor/year. Transmutation of neptunium, as shown in Figure 4follows a similar trend to that of americium. For neptunium for the 5% and 100% cases respectively 91% and 40% of the initial neptunium is transmuted at average rates of 0.8 and 12.5 kg/reactor/year.

The decay time does have a significant impact on the amount of curium that is produced. For the case with 100% density in the centre pin, there is only 35% as much mass of curium produced for the 3 month decay case relative to the 2 year decay case by the 4^{th} irradiation cycle, and 16 kg total less curium produced.

The addition of the minor actinides and lanthanides to the centre pin of the fuel bundle serves to lower the coolant void reactivity (CVR). The reduction in CVR from standard natural uranium (NU) fuelled CANDU reactors for each density is shown in Figure 5. The reduction in CVR ranges from 0.5 to 1.0 mk for the 5% density and 7 to 8 mk for the 100% density.

During this fuel cycle plutonium is produced in the center pin from the decay of curium isotopes. Subsequent to the four irradiation cycles and a final decay period the pin can be processed to recover and recycle the generated plutonium.

These calculations indicate that a very efficient destruction of Am and Np can be achieved using the unseparated fraction of minor actinides and lanthanides. The latter provides a major simplification in the used fuel separation process. At the same time, this burning scheme has the significant added benefit of providing a reduction in the coolant void reactivity without any significant performance loss in the reactor efficiency for producing electricity.

3. Homogeneous Method, Full Core of Am/LEU

This scenario involves a different reprocessing scheme than that in the previous case; in this case americium only is separated out of the used nuclear fuel. There have been some recent advances in Am-only separation [8], [9].

For these simulations the isotopic composition is given in Table 2. The separated americium was then modeled as mixed with recycled uranium (RU). This option utilizes the uranium from the used fuel, as well as transmuting the americium [10], [11]. Americium acts as a neutron poison so extra U-235, above that in natural uranium, which is the nominal fuel for CANDU reactors, was required to compensate for the loss of reactivity. The enrichment of the RU was also required to be varied by blending with SEU, in order to maintain a constant burnup of either 7.5 MWd/kg(IHE) or 21 MWd/kg(IHE). A CANFLEX fuel bundle design was used for this study, see Figure 2. These lattice cell calculations were performed using WIMS-AECL [5].

Table 2. Input isotopic composition of the Am from used nuclear fuel

Isotope	% by Weight
Am-241	73.8

Am-242m	0.12
Am-243	26.1

Four different CANDU reactor cases will be discussed here. Two different burnups were studied, the nominal burnup for a CANDU-6 fuelled with natural uranium, 7.5 MWd/kg(IHE), and a higher burnup, 21 MWd/kg(IHE). These cases are:

1. 0.28% Am, with a burnup of 7.5 MWd/kg(IHE). The centre pin was 2.4% Dy in zirconia,
2. 0.28% Am, with a burnup of 7.5 MWd/kg(IHE). The centre pin was 7% Am in zirconia,
3. 0.28% Am, with a burnup of 21 MWd/kg(IHE). The centre pin was 1.3% Dy in zirconia,
4. 0.28% Am, with a burnup of 21 MWd/kg(IHE). The centre pin was 3.7% Am in zirconia.

The results for the four cases are given in Table 4. These four cases have a neutron poison in the centre pin of the fuel bundle in order to lower the CVR. Cases 2 and 4, with Am in the centre pin, enable the maximum amount of Am transmutation (2067 and 2854 g/tonneIHE) although the fraction of Am transmuted is the lowest (45% and 77%) for a given burnup. The reduction in the fraction of Am transmuted is less significant at the higher burnup, 77% versus 79%. Up to 20.7 GWe of primary LWR power can be supported for one GWe of CANDU reactor power (case 2). However, it should be noted that the fraction of Am transmuted in this case is lower. For the higher transmutation fraction cases (cases 3 and 4), up to 6.1 GWe of primary PWR can be supported.

Table 3. Parameters for the four Am/LEU cases

Case	Model	Input Amount of U-235 (wt%)	Burnup (MWd/kg(IHE))
1	0.28% Am, 2.4% Dy	1.01	7.5
2	0.28%, 7%Am in centre	1.04	7.5
3	0.28% Am, 1.34% Dy	1.34	20.9
4	0.28%, 3.7% Am in centre	1.36	21.1

Table 4. Results for the Am/LEU cases.

Case	Model	Input Am (g/tonne IHE)	Net Am Transmutation		Support Ratio (GWe primary PWR: GWe burner reactor)
			Change (%)	Mass Transmuted (g/tonne IHE)	
1	0.28% Am 2.4% Dy	2843	-53.3	1516	13.0
2	0.28%, 7% Am in centre	4522	-45.7	2067	20.7
3	0.28% Am, 1.34% Dy	2841	-79	2235	4.6
4	0.28%, 3.7% Am in centre	3726	-77	2854	6.1

For case 4, the decay heat is reduced 40 years after discharge, and a decrease of more than 60% is seen after 1000 years, Figure 6. The decay heat reduction was calculated relative to two scenarios, the first scenario is a reference case with no recycling of Am into the CANDU reactor. The second case is the case 4 recycle scenario in which Am is separated out from the PWR used fuel, with the rest of the spent fuel going to a repository. The Am is recycled into the CANDU reactor, and the CANDU spent fuel goes into the repository. The Am mass balance is taken into account in this calculation. For case 4, one GWe of CANDU reactor power can support 6.4 GWe of LWR reactor power.

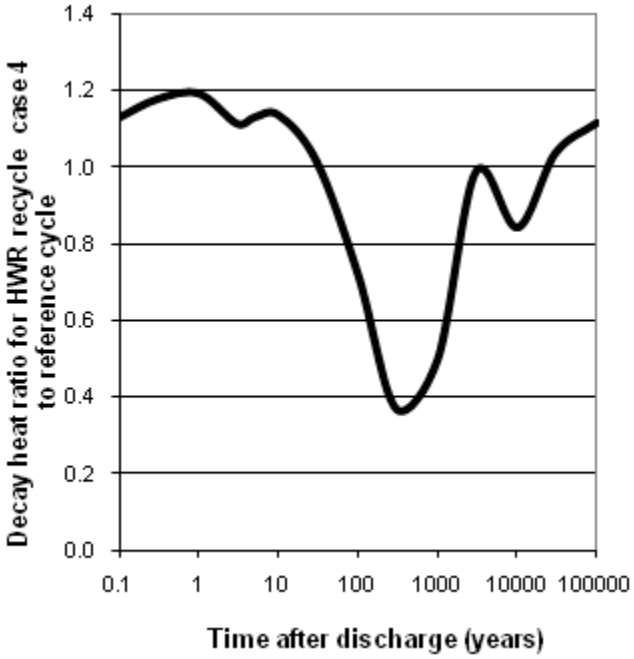

Figure 6. The ratio of decay heat relative to the reference case with no Am recycle to the scenario case 4

4. Group-Extracted TRU-MOX in CANDU

In this case, a group-extracted reprocessing scheme is assumed for the transuranic elements. All of the transuranic elements are used: Pu, Np, Am, and Cm, and their relative amounts are kept the same as in the original LWR used fuel.

The input fuel simulated in this study was used nuclear fuel from a LWR that had been cooled for 30 years and then reprocessed to recover all the TRU [11]. The cooled TRU were then mixed with natural uranium to form a MOX fuel. This fuel was then irradiated in a CANDU 6 reactor. The input isotopic composition of the fuel is given in Table 5.

Table 5. Input isotopic composition of the transuranic nuclides.

Isotope	% by weight	Isotope	% by weight
Np-237	4.7	Am-241	9.9
Pu-238	1.3	Am-243	0.76
Pu-239	59.2	Cm-243	0.001
Pu-240	20.1	Cm-244	0.072
Pu-241	3.0	Cm-245	0.12
Pu-242	3.8	Cm-246	0.001

The fuel is 4.0% TRU by weight (heavy elements). The fuel bundle design used was the 43-element CANFLEX design. This is the same geometry as shown in Fig. 2, but the materials are different in this case. The centre pin has a composition of dysprosium in a zirconia matrix in order to reduce CVR.

Lattice cell calculations were performed using WIMS-AECL [5] and full-core modelling used the RFSP computer code [13]. The exit burnup of the fuel was 43.4 MWd/kg(IHE). This gave a total TRU transmutation of 42%. The percentage of each isotope transmuted and the mass transmuted per year are given in Tab. 10. Note for this table a positive value is a destruction and a negative value is a creation. There is an increase in curium mass, but it should be noted that most curium isotopes are short-lived and contribute to the decay heat on the same time scale as the fission products.

The evolution of the transuranic elements is shown in Figure 7. The high neutron economy of the CANDU reactor gives high values of destruction for the fissile nuclides. The fissile content of the TRU vector, initially 63%, drops to 30%. The support ratio for this fuel cycle is 11.2. This reduction in actinides corresponds to a reduction of the decay heat of the used fuel by about 40% at 1000 years exit compared to LWR used fuel that is not reprocessed and transmuted in a CANDU reactor. This could have significant impact on the capacity of a geological repository.

Full-core calculations of a TRU-MOX core have been completed to demonstrate the feasibility of fuelling a CANDU 6 core with this fuel. Several changes are needed with respect to the natural uranium (NU) fuel cycle. The fuelling scheme for NU generally involves replacing eight bundles in a fuel channel in each fuelling operation, referred to as an eight-bundle shift. Due to the higher initial reactivity of the MOX bundle, in order not to introduce too much localized reactivity into the core, the fuelling scheme is reduced to a combination of one- and two-bundle shifts, applied to 264 and 116 channels, respectively. A detailed study on the impact on fuelling machine utilization has not been completed, but the higher burnup and therefore longer dwelling time of the fuel compensates for the smaller number of bundles shifted during refuelling. This results in a refuelling rate of about 3 bundles per day, compared to 15 bundles per day in the natural uranium CANDU 6 reactor.

A time-average model of the core was created to examine the maximum channel and bundle powers. In addition to this model, an instantaneous snapshot model was also examined. The instantaneous model, which generates random ages for the fuel channels, was used to analyze the power increase that occurs when refuelling the reactor, referred to as the refuelling ripple. Values for the maximum channel and bundle powers are given in Table 7. As is expected, the maximum channel and bundle powers are higher for the instantaneous model, which accounts for an increase in power as fresh fuel bundles are added to the core upon refuelling. These values are all within normal CANDU reactor operating conditions.

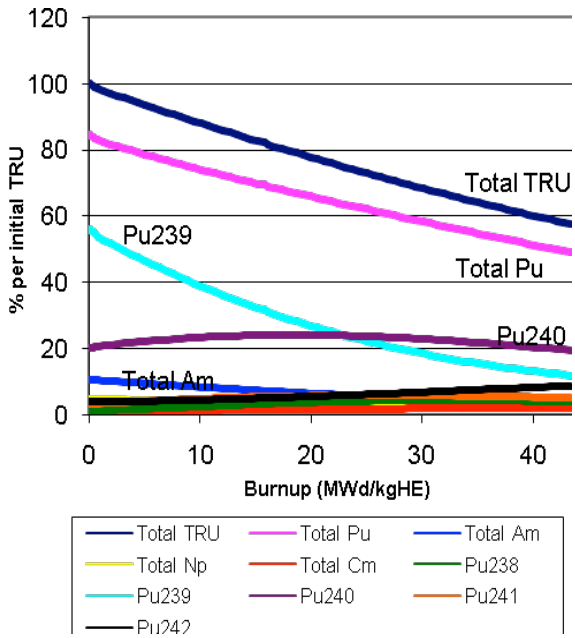

Figure 7. The isotopic evolution of TRU during transmutation in the CANDU reactor.

Table 6. Transmutation of TRU in the TRU-MOX scenario

Nuclide	% Transmuted	Mass Transmuted (kg/year)
Np-237	51.0	18.9
Total Np	48.1	17.9
Pu-238	-206.6	-21.2
Pu-239	78.6	348.4
Pu-240	2.4	3.8
Pu-241	-68.8	-16.5
Pu-242	-128.2	-38.4
Total Pu	41.1	276.8
Am-241	84.2	65.8
Am-243	-221.1	-13.3
Total Am	62.2	52.4
Cm-242		-6.1
Cm-243	-2774.4	-0.22
Cm-244	-1676.5	-9.5
Cm-245	-158.6	-0.15
Cm-246	-1706.0	-0.13
Total Cm	-2374.4	-16.1
Total TRU	41.9	330.2

Table 7. The maximum channel and bundle powers for the time-average and instantaneous models for the MOX fuel cycle

	Type of Model	
	Time-Average	Instantaneous
Maximum Channel Power (kW)	6300	7200
Maximum Bundle Power (kW)	820	910

5. Heterogeneous Core Method, Target Channels of Am and Cm

In this scenario a CANDU 6 core is fuelled with 0.9% fissile RU and peripheral channels around the outside of the reactor (30 channels total) are fuelled with Am/Cm in an inert matrix (Figure 8)[4]. The isotopic composition of Am and Cm in spent fuel used in the analysis was taken from the used fuel database maintained by the Nuclear Energy Agency and is shown below. The data set used is Takahama-3 47.03 GWd/MT(IHE). The used fuel was decayed for 30 years, and then the Am and Cm were separated out. The burnup achieved for the RU is 12.2 MWd/kg(IHE).

The peripheral channels are selected for the Am/Cm transmutation in this analysis because of their minimal effect on the reactor operation in a CANDU 6 reactor. The portions of Am/Cm in the inert matrix fuel (IMF) mixture are 14%, 19%, 26%, and 35% by weight. Silicon carbide (SiC) has been used as the IMF material in this model. The choice of the inert matrix is not important at this stage; any material that is transparent to neutrons is suitable from the standpoint of the physics calculations performed here. If this fuel cycle were to be developed further, a study would be done to determine the most suitable inert matrix.

Table 8. Isotopic composition of the americium and curium in the central pin.

Nuclide	% by weight
Am-241	85.7040
Am-242m	0.0666
Am-243	12.0214
Cm-242	0.0002
Cm-243	0.0268
Cm-244	1.7555
Cm-245	0.3783
Cm-246	0.0465
Cm-247	0.0007

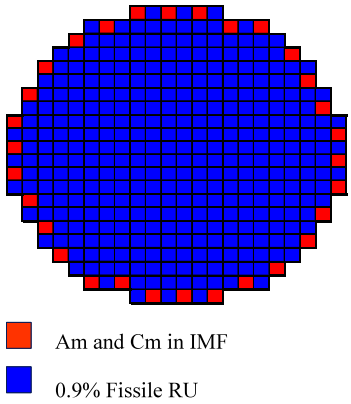

Figure 8. Schematic diagram of the Am/Cm target model for a CANDU 6 core

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Four different bundle designs were modelled for the Am/Cm carrier. The bundle designs are 21-element, 24-element, and 30-element bundles and the 43-element CANFLEX bundle.

Figure 9 shows the relationship between the support ratio (SR) and the destruction of americium using a 30-peripheral channel loading scheme. A support ratio of 4 means that americium produced by 4 GWe of LWR reactors can be loaded into 1 GWe of CANDU 6 reactors. This plot is for the CANFLEX fuel bundle but the relationship is the same for all of the bundle designs. A lighter fuel bundle has a shorter residence time to achieve the same percentage of Am/Cm transmutation as that for a heavier bundle. However, with a lighter fuel bundle, less mass of Am/Cm can be fuelled in the reactor.

The graph in Figure 10 shows the link between the percentage of Am that can be transmuted and the support ratio, and how the heavier bundles require a longer residence time to achieve an equivalent destruction. If a high support ratio is desired, then a lower destruction rate is obtained; conversely if a high percent transmutation of Am is desired then more GWe of CANDU reactors are required. Support ratios, for once-through applications in fuel cycles, serve as an indication of how much minor actinides are loaded into the reactors in the various scenarios. An effective strategy to burn Am (and other minor actinides) would need to balance throughput and the actual quantity of MA that is transmuted.

A heavier bundle allows more Am to be input in to the CANDU reactor at one time, but a longer irradiation time is required to achieve the same Am/Cm destruction. Therefore for a chosen destruction and support ratio the residence time can be chosen by selecting a lighter or heavier fuel bundle design.

Varying the initial amount of Am/Cm in the bundle produces the same relationship as shown in Figure 10, see Figure 11. The amounts of Am/Cm that were modelled are 14%, 19%, 26%, and

35% by weight. Figure 9 shows the effect of residence time on the destruction of Am/Cm and on the support ratio for the case with 19% initial concentration of Am/Cm. The calculations varying the initial concentration are for the CANFLEX fuel bundle design only. This provides another means to choose the fuel design, whereby the initial Am/Cm concentration of the bundle can be chosen for a particular fuel bundle, support ratio/% destruction, and residence time.

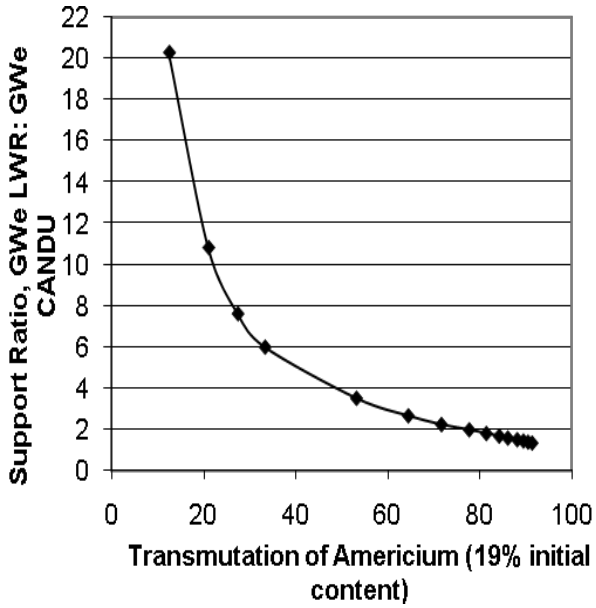

Figure 9. Relationship between the support ratio and the transmutation of Am.

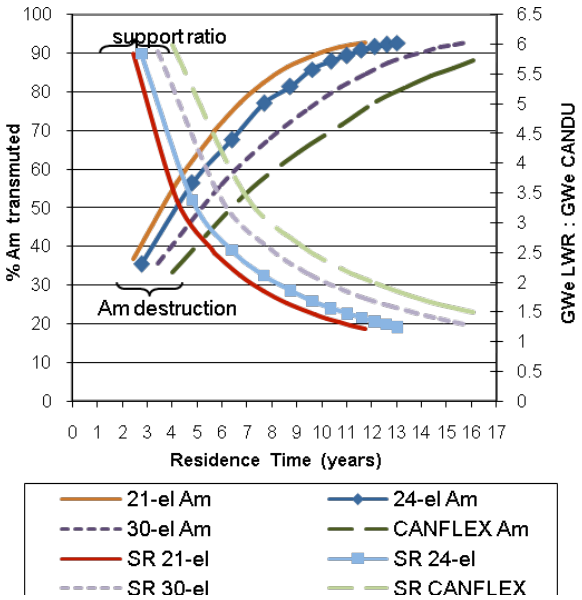

Figure 10. Transmuation of Am and the support ratio vs. residence time for each of the four different fuel bundle designs

During the irradiation, there is a growth of plutonium initially from the alpha decay of Cm-242 to Pu-238, and from the electron capture decay of Am-242 to Pu-242. The isotopic evolution of the Am/Cm is shown in Figure 12. The figure shows the results for the 21-element fuel bundle with an initial composition of 19% Am/Cm (by volume). Note that for any other case, the trends are the same, but the scales on the axes will stretch/contract.

The method of using 30-periphery channels for Am/Cm transmutation could be used if minimal operational change is desired. Due to lower neutron flux in the outer channels in the CANDU 6 reactor, the power contribution from these channels to the total power is small compared to the channels in the centre of the core. This 30-periphery channel method will also maximize the utilization of neutrons that would escape from the core otherwise. However, the trade-off between percent transmutation and support ratio can result in low actinide destruction: less than 10% transmutation rate per year even at low loading using 21-element bundle at 19% concentration. To achieve a higher transmutation rate of ~50%, the support ratio reduces to 3.5

for all four bundle types, that is roughly equivalent to ~35 kg of actinide destruction per year. Future work will include calculations of the impact of these scenarios on the decay heat. This has not been performed at this time due to limitations in the codes.

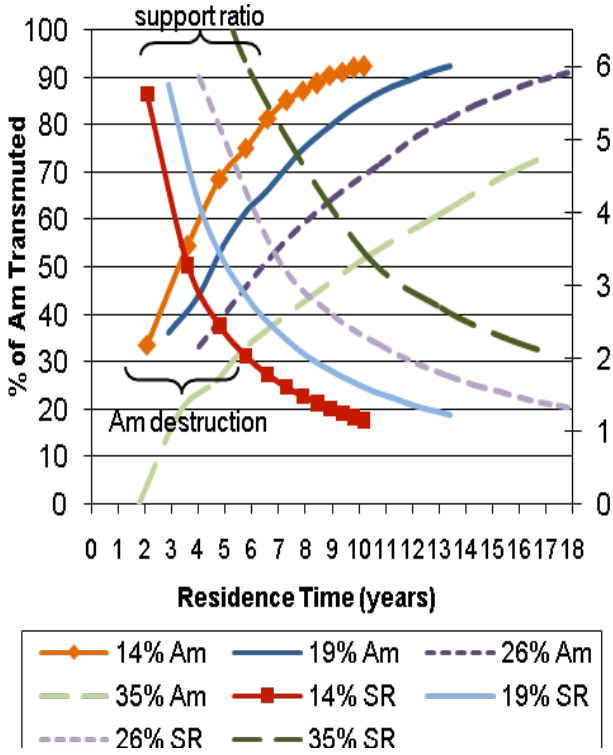

Figure 11. Transmutation of americium and the support ratio vs. residence time for each of the four different initial amounts of Am/Cm.

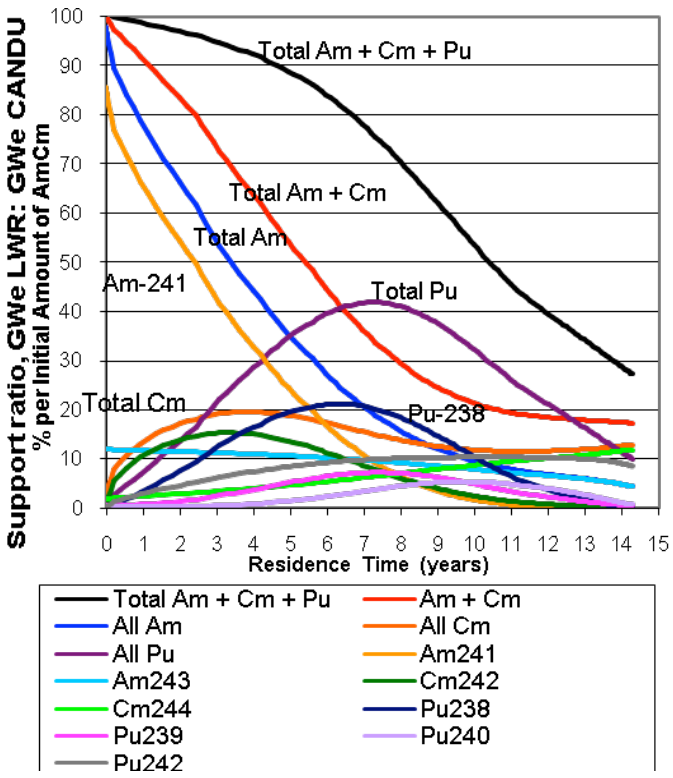

Figure 12. Isotopic evolution of the significant transuranic nuclides in the Am/Cm fuel for the 21-element fuel bundle with an initial composition of 19% Am/Cm.

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6. Conclusions

In this paper, several different actinide transmutation schemes in a CANDU reactor were presented. These methods are classified under two categories: heterogeneous and homogeneous actinide loading in CANDU reactors. In the former method, actinides are confined to target pins or bundles and loaded in the reactor separately from the rest of fuel. In the homogenous methods, actinides are homogeneously mixed with fuel and placed in all channels in the reactor.

The advantages of these methods are that they require minimal operational and fuel changes in existing CANDU reactors. In the case of using the centre pin of the bundle to carry the actinides, the concept is very similar to the "low void reactivity fuel" (LVRF) CANDU fuel design that contains a centre neutron absorber and slightly enriched uranium in the remaining fuel pins. In the heterogeneous channel method where actinide target bundles are placed in dedicated transmutation channels of the reactor, the periphery channels are selected to minimize the operational effect while utilizing neutrons in the reflector region. On-power refuelling capability

of a CANDU reactor allows the residence time of actinide targets to differ significantly from fuel bundles. With high neutron fluence, very high destruction of the actinides can be achieved. The disadvantage of this scheme is that the total amount of actinide destruction is limited since only small part of the reactor is being used for actinide transmutation. The heterogeneous methods are suitable when there are sufficient numbers of existing CANDU reactors to support a LWR fleet.

Two different homogeneous actinide fuels are presented in this paper corresponding to different actinide partitioning schemes: separated Am mixed with LEU, and group-extracted actinides mixed with natural uranium. Significant actinide mass destruction can be achieved using whole core loading of these homogeneous actinide fuels. The actinide transmutation in a single CANDU reactor is in the order of several kilograms per year, achieving a support ratio (LWR to CANDU power) of between 11~20 depending on the actinide-partitioning scheme. The limitation in the homogeneous method is that the fuel and targets are subject to the same neutron fluence, and therefore this method does not have the flexibility of the heterogeneous core scenarios. The discharge actinide fuel may require secondary reprocessing and transmutation to achieve high destruction comparable to the heterogeneous methods. A higher fuel burnup will increase the actinide destruction fraction.

Several different strategies are being studied to maximize the CANDU reactor utilization for actinide destruction. The currently existing CANDU reactors can process small quantities of actinides without significant operational changes. This paper has not examined actinide destruction using thorium as a fuel matrix, rather than uranium. Thorium is anticipated to be a better transmutation matrix than uranium because fewer higher mass actinides would be produced during irradiation and valuable U-233 would be produced for subsequent recycle.

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