A Study on the Optimal Cooling Time of Actinide Feedstock for use in Transuranic Mixed Oxide Fuels in CANDU Reactors

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Abstract

Transuranic actinides from spent fuel can be burned in current reactors as a mixed oxide (MOX) with natural uranium, reducing spent fuel radiation and heat loads. Detailed reactor physics analysis of transuranic MOX in CANDU (TRUMOX-CANDU) showed significant actinide conversion while maintaining operational performance. The different cooling times of spent fuel feedstock affect the MOX fuel actinide composition, which: i) alters the neutronics of the resulting TRUMOX fuel, ii) affects actinide conversion, and iii) determines end of cycle composition, dictating spent fuel radiation and heat loads. These effects are examined to find a balance between in-core performance, actinide conversion and end of cycle spent fuel properties. TRUMOX fuel compositions, dictated by the cooling time of the actinide feedstock, are evaluated with WIMS-AECL CANDU lattice cell calculations. In-core neutronics, actinide conversion ratio, and end of cycle fuel composition are examined to estimate an optimal cooling time for the actinide feedstock before use in TRUMOX fuel.

1. Introduction

A major issue facing the nuclear industry today is the management of spent fuel. By employing reprocessing strategies, the liability of spent fuel stockpiles can be turned into a commodity. The use of reprocessing can reduce long term waste issues and help to close the nuclear fuel cycle. Reprocessing seeks to transmute long lived isotopes present in spent fuel (i.e., actinides) and leave only the short-term (< 500 y) by-products (i.e., fission products). The fission products are the actual waste product of the fuel cycle and can be vitrified and placed in a disposal facility. The actinides are made up of two streams, uranium and Np + transuranics (i.e., Pu, Am and Cm), referred to in this paper as transuranics. The uranium stream is separated out from the other actinides and can be re-used directly in thermal reactor systems through re-enrichment, via blending with newly mined uranium, or in mixed oxide formats. The transuranics can be separated out by element or can be group extracted from the spent fuel. The transuranics can then be transmuted to shorter lived isotopes through neutron capture or fission. Transmutation of the transuranics can be accomplished through the use of targets in fast or thermal reactors or can be blended into mixed oxide fuels to be burned in thermal reactor designs. Mixed oxide fuels blend uranium (recycled, natural or depleted) with the transuranics providing a path for transmutation/fission in current thermal reactor designs. The remaining actinides can be cycled through other thermal and fast systems until they are fully transmuted. Partitioning the spent fuel into fission products, uranium and transuranics and then utilizing the latter two components in further fuel cycles, removes the uranium and long lived transuranics from the waste stream. This

serves to reduce the longevity of the resultant waste material (fission products only) from 100,000 years to about 500 years (the time for fission products to decay below the level of radiotoxicity of naturally occurring uranium ore). There are also a few long lived fission product isotopes (half lives of > 200,000 y) which would remain present in the waste stream. However, the activity of these long lived isotopes is below the level of radiotoxicity of naturally occurring uranium ore and thus has limited impact on longevity concerns when compared to the actinides. Research into the partitioning and transmutation of long lived fission products is being conducted but this is outside the focus of this paper and is not explored herein.

Burning transuranics from spent fuel in MOX format in current thermal reactor designs provides a suitable intermediary step before moving to fast reactors to complete the transmutation process. Such a multi-stage approach reduces the burden on fast reactor technologies and utilizes current proven reactor designs. Fast reactor systems have increased complexity, less widespread use and deployment than thermal systems, and have met with limited operational success. MOX fuels for thermal reactors can blend extracted transuranics (either group extracted or separated) with uranium (either recycled or newly mined) or thorium. The group extracted option is simpler from the reprocessing side and blending group extracted transuranics with natural uranium (NU) is an area of developing research [1][2][3]. This research has been the subject of extensive study beginning with the reprocessing of excess nuclear weapons material including studies on plutonium disposition in CANDU [4][5]. MOX fuels composed of blended plutonium and uranium are in use in currently operating pressurized water reactors (PWRs) in France, Belgium, Switzerland and Japan with core loadings of 10-20% MOX. Detailed feasibility studies including full core physics simulations of transuranic mixed oxide (TRUMOX) fuels have been performed for CANDU systems (including both 380 and 480 channel designs) and the results show appreciable actinide conversion with safe operation within the standard envelope [6][7][8].

Previous TRUMOX studies included multi-stage physics simulations using lattice cell calculations in WIMS-AECL [9] and 3D supercell calculations in DRAGON [10] to produce a full core diffusion model of a TRUMOX fuelled CANDU-900 in RFSP-IST [11]. The first stage is the specification of a fuel configuration with a mixed oxide composition in advanced bundle geometry which is determined through successive lattice simulation. Following the definition of a suitable fuel configuration meeting burnup and actinide targets, incremental cross-sections for CANDU control devices (e.g., liquid zone controllers, adjusters, absorbers and shutdown rods) interacting with the lattice cell were computed using 3D supercell calculations in DRAGON. These calculations were used to generate full burnup homogenized macroscopic lattice cell cross-sections and the set of homogenized incremental cross-sections from the super-cell calculations for all control devices. These values are utilized by the full core diffusion code, RFSP, to compute a 3D full core diffusion solution (RFSP fuel tables were generated from the WIMS-AECL lattice model using the WIMS-Utilities code to produce micro-depletion tables).

The diffusion model simulates the full core accounting for internal structural components and control devices using the homogenized lattice and super-cell cross-sections as building blocks to reproduce the core configuration and solve the diffusion equation to determine the neutron flux distribution within the core. The online fuelling pattern (specific irradiation levels, fuelling scheme and frequencies to balance the desired fuel burnup level with the maintenance of the

proper core reactivity, power levels and flux profiles) is adjusted to produce an optimal "equilibrium" core configuration lying within the standard CANDU operational envelope. The calculations include the responses to changes in conditions (e.g., temperature, moderator composition, etc.) and the full core responses to the different control devices. The optimal equilibrium or "time average" model is further refined using instantaneous snapshots of the core with random channel ages to ensure the model remains within the desired operational envelope. Specifically, the online fuelling nature of the CANDU reactor means that the core at each instant in time will show small deviations from the time-average conditions described earlier. The online fuelling requirements of CANDU reactors put specific demands on the control system and fuelling events must be studied in detail to ensure that the reactor remains within the operational envelope during fuelling. The finalized model can then be evaluated for normal and accident operations to determine the response of the system and ensure acceptable behaviour under the defined conditions.

Detailed descriptions of the full core modelling process as applied to TRUMOX fuels in CANDU-900 are available in Reference [7] along with information on the fuel configuration and evaluated full core model. The full core model simulates the 480 channel CANDU-900 (900 MWe output) reactor with 13 fuel bundles in each channel (similar to the Bruce and Darlington units). The reference fuel of the CANDU-900 to which the TRUMOX fuel is compared is the 37 element NU fuel. The NU fuelled CANDU-900 is referred to as NU-CANDU-900 while the TRUMOX fuelled CANDU-900 is referred to as TRUMOX-CANDU-900. More detailed assessments of this full core model are performed in References [12][13], focusing on assessing the ability of the combined reactor/fuel system to maintain a standard operational envelope, including effects of coolant voiding, short-term fuelling simulations, fuel ramp assessments, flux mode analyses and core responsiveness. Additionally, a study into the effects of variations in fuel composition simulating the blending of different actinide feedstock on the lattice physics has been performed [14]. This study looked at the effects of fuel composition variations on lattice cell reactivity, burnup, actinide conversion and coolant voiding and provided insight on the tolerances required in blending processes used to generate a desired fuel composition to ensure that desired neutronic characteristics and actinide conversion and burnup targets are maintained.

The TRUMOX CANDU system is being explored because of the capability to utilize the transuranic actinides from spent fuels and provide an intermediary step for transmuting these long lived isotopes before moving on to fast reactor systems. Thus current inventories of spent fuel become a commodity and provide actinide feedstock for the manufacture of TRUMOX fuels. Spent fuel has been amassed over decades of operation of reactors and as a result has a wide array of initial enrichments (fresh fuel U-235 content), irradiation histories (i.e., burnup), and cooling times. Blending processes can allow the manufacture of a specific fuel composition (within a certain tolerance) to match a desired fuel and reactor configuration. However, it is beneficial to also assess an optimal cooling time for spent fuel before it is employed as a transuranic actinide feedstock. As the spent fuel is left to cool, decay reactions cause changes in the isotopic composition which alters the proportions of transuranic actinides available as a feedstock to the production of TRUMOX fuel. This variation in actinide composition: i) alters the neutronics of the resulting TRUMOX fuel, ii) effects the actinide conversion of the fuel

cycle, and iii) determines the end of cycle composition which dictates the spent fuel radiation and heat loads. By examining these effects, a balance between in-core performance, actinide conversion and end of cycle spent fuel properties can be explored. In this study, the TRUMOX fuel compositions dictated by the various cooling times of the actinide feedstock are evaluated with WIMS-AECL calculations for a CANDU lattice cell over a full burnup cycle. These calculations examine the in-core neutronics, actinide conversion ratio, and the end of cycle fuel composition. Through the study of these parameters, an optimal cooling time for the actinide feedstock before use in TRUMOX fuel can be estimated. The lattice cell model used is discussed in Section 2 along with the standard TRUMOX-CANDU-900 fuel composition and the PWR spent fuel feedstock used for this aging study. Exploration of the effects of using spent fuel with different cooling times as actinide feedstock on lattice cell physics, actinide conversion and end of cycle composition are discussed in Section 3 with conclusions presented in Section 4.

2. Fuel Composition and Lattice Cell Model

This study begins with the reference lattice cell and fuel bundle configuration from the TRUMOX-CANDU-900 [7]. The lattice characteristics are consistent with a CANDU design (i.e., horizontal fuel channel, pressure tube, calandria tube, heavy water moderator and coolant, etc.). The TRUMOX design uses an advanced fuel bundle design with 43-element fuel bundle with a central pin composed of dysprosium-zirconium-oxide (67 wt% Dy_3O_2) [7] burnable neutron absorber (BNA). The central absorber pin is a larger diameter (17.4 mm) compared to the remaining 42 fuel elements of the bundle which are smaller diameter (11.4 mm). These elements are arranged in concentric rings of 7, 14, and 21 elements surrounding the central BNA element and are fuelled with a blend of natural uranium and transuranic actinides.

The TRUMOX-CANDU-900 lattice cell is compared to the standard NU-CANDU-900 lattice cell in Figure 1 [12]. The NU-CANDU-900 uses a 37 element bundle (all element diameters are 13.1 mm), which is fuelled entirely with natural uranium (fresh fuel fissile content of 0.71 wt%, entirely U-235) and has a target burnup of 9,000 MWD/T in the CANDU-900 reactor design.

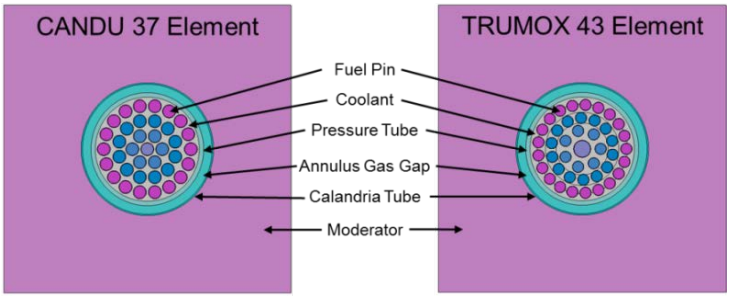

Figure 1 2D Lattice Cell Models for 37-Element CANDU and 43-Element TRUMOX [12]

2.1 Reference TRUMOX Fuel Composition

The TRUMOX reference fuel [7] is a blend of group-extracted transuranic actinides and natural uranium. This mixed oxide fuel is used in the 42 fuel elements (the remaining centre element being a BNA) of the TRUMOX 43-element bundle in the CANDU-900 reactor. The transuranic

actinide composition (Np, Pu, Am and Cm) used in the reference study is dictated by the isotopic composition of a specific spent fuel feedstock. For the reference TRUMOX fuel, the transuranics are sourced from cooled spent fuel from a prototypical PWR design with an irradiation of 45,000 MWD/T of heavy element and a cooling time of 30 years. The isotopic composition of the transuranic mixture is dictated primarily by these parameters of irradiation history and cooling time. The reference transuranic actinide composition is based on data from studies at Oak Ridge National Laboratory that predicts the probable yields of actinides from spent fuel reprocessing. The transuranic mixture contains 12 isotopes with the composition shown in Table 1 [7][15].

Table 1 Reference Transuranic Actinide Composition for TRUMOX-30 Fuel [7]

#	Isotope	wt%	#	Isotope	wt%	#	Isotope	wt%
1	Neptunium-237	4.698	5	Plutonium-241	3.040	9	Curium-243	0.001
2	Plutonium-238	1.301	6	Plutonium-242	3.800	10	Curium-244	0.072
3	Plutonium-239	56.243	7	Americium-241	9.907	11	Curium-245	0.012
4	Plutonium-240	20.099	8	Americium-243	0.763	12	Curium-246	0.001

The TRUMOX-CANDU-900 reactor has been designed to meet a burnup target of 30,000 MWD/T heavy element through the blending of the transuranic actinide mixture with natural uranium (NU, fissile content of 0.71 wt% from U-235). This specific fuel composition is called TRUMOX-30 and is 96.9% NU and 3.1% transuranic actinide mixture. The resultant mass of fuel is 17.38 kgHE per bundle with a fresh fuel fissile content of 2.53 wt% (including Pu-239 and Pu-241 from the transuranics and U-235 from the NU). In the reference TRUMOX-CANDU-900, this fuel achieved an average burnup of 29,980 MWD/T (29.980 MWD/kg) and resulted in an actinide conversion of approximately 35%. The initial and final concentrations for the TRUMOX-30 fuel are provided in Table 2 (these burnup values are calculated using the ENDF-B-VI libraries with patches for curium and dysprosium [7]).

Table 2 TRUMOX-30 Actinide Conversion (Burnup = 29,980 MWD/T of heavy element) [7]

Conc. (g/bundle)	U-235	Np Total	Pu Total	Am Total	Cm Total	Total Actinides
Initial	119.88	24.43	439.23	55.48	0.45	519.58
Final	24.55	13.47	293.09	20.80	11.62	338.99
Final-Initial	-95.34	-10.96	-146.13	-34.68	11.17	-180.59
% change	-79.53%	-44.86%	-33.27%	-62.51%	2491.04%	-34.76%

2.2 Fuel Aging Study Transuranic Actinide Feedstock

For this study, a comparison is made between the transuranic actinide compositions sourced from the same spent fuel feedstock that is cooled for different times. The original fuel for this study was a prototypical PWR fuel with an initial enrichment of 4.5 wt% U-235. The fuel configuration is irradiated in a PWR infinite pin cell (simulated with WIMS-AECL) up to a burnup of 45,000 MWD/T. Following this, various cooling times (from 5-40 years) for the spent

fuel were used to calculate the changes in isotopic composition using the ORIGEN code. The isotopic composition of the transuranic actinide mixture changes over the length of the cooling time due to radioactive decay and transmutation. The evolution of the isotopic content of the transuranic actinide mixture over the cooling time from 5 to 40 years is shown in Figure 2. The numeric isotopic content for the various cooling times is also provided in Table 3.

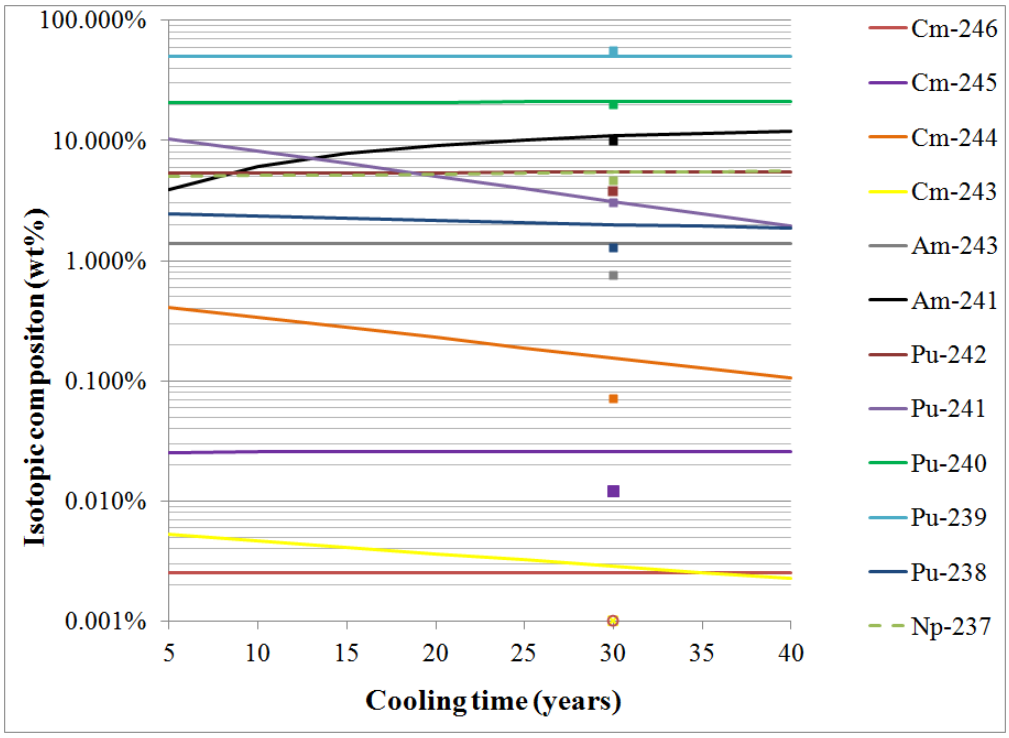

Figure 2 Aging Study Evolution of Isotopic Composition over the Cooling Period

Table 3 Aging Study Transuranic Actinide Compositions for 5 to 40 year Cooling Periods

Cooling Time (years)	5	10	15	20	30	40
Isotope	**Wt%**	**Wt%**	**Wt%**	**Wt%**	**Wt%**	**Wt%**
Np-237	5.076	5.122	5.183	5.255	5.425	5.618
Pu-238	2.429	2.338	2.250	2.167	2.007	1.858
Pu-239	50.317	50.366	50.420	50.466	50.555	50.642
Pu-240	20.675	20.759	20.825	20.882	20.978	21.043
Pu-241	10.376	8.159	6.415	5.044	3.118	1.927
Pu-242	5.392	5.398	5.404	5.410	5.421	5.431
Am-241	3.917	6.110	7.813	9.134	10.925	11.959
Am-243	1.380	1.380	1.381	1.382	1.384	1.385
Cm-243	0.005	0.005	0.004	0.004	0.003	0.002
Cm-244	0.405	0.335	0.277	0.229	0.156	0.107
Cm-245	0.025	0.025	0.025	0.025	0.025	0.026
Cm-246	0.003	0.003	0.003	0.003	0.003	0.003

The reference TRUMOX transuranic actinide composition (shown previously in Table 1) is displayed in Figure 2 for comparison purposes as single data points at 30 years cooling. The aging study mixture composition is different from the TRUMOX reference mixture derived from the Oak Ridge studies. This is due to differences in irradiation history (i.e., duration, power level, location in the PWR core) and initial fuel enrichment before irradiation in the PWR. There may also be some variation due to WIMS-AECL PWR pin cell modelling. The TRUMOX reference composition is used to provide perspective but the primary comparison in this study is between the isotopic contents of the transuranic actinide mixtures of the spent fuel with different cooling times. The evolution of the isotopic composition of the transuranic actinides over the cooling time (5-40 years) shown in Figure 2, displays several trends. Of the 12 transuranic isotopes, the 5 that change significantly over the cooling time are shown in Figure 3.

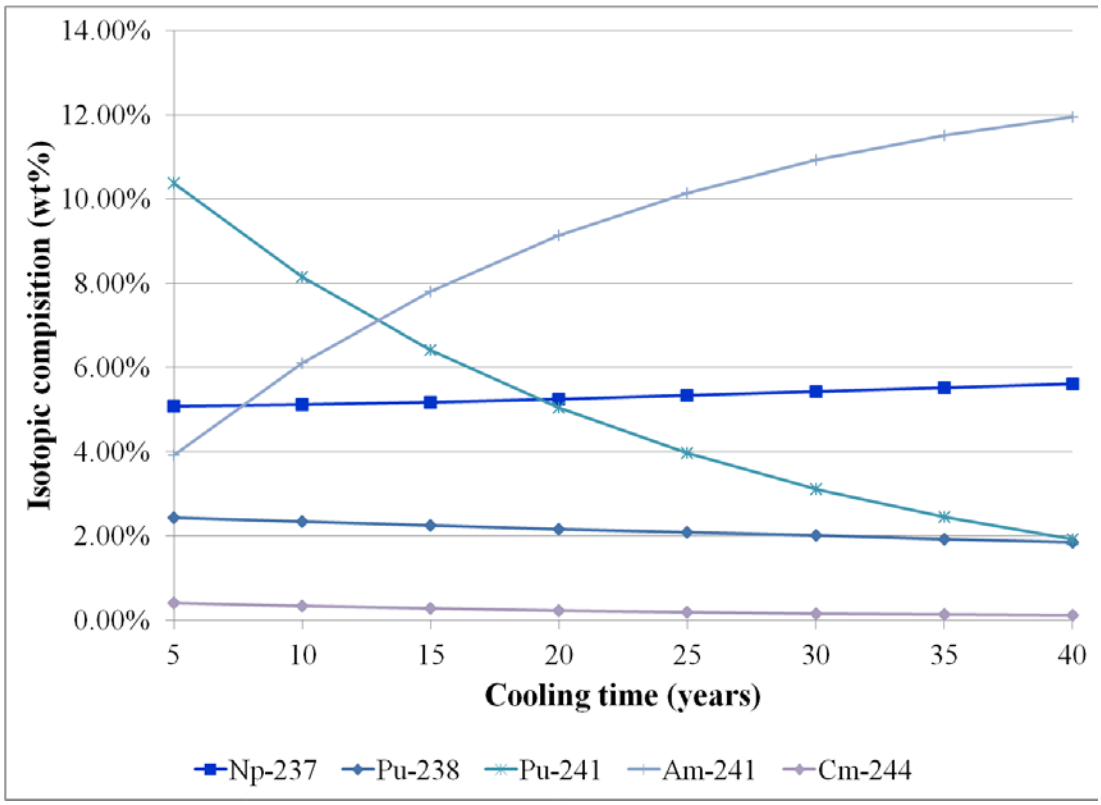

Figure 3 Aging Study Isotopes that Change Significantly over the Cooling Period

The most significant change in isotopic content during the cooling period is the decay of Pu-241 into Am-241 (with a half-life of about 14.36 years), shown in Figure 3). This is important as Pu-241 is a fissile isotope while Am-241 with its large neutron-capture cross section acts as a burnable absorber within the fuel. Thus, as the fuel is cooled, the fissile content decreases and the neutron-capture of the fuel increases (due to the increased Am-241 content, which has a high capture cross section). This means that more actinide mixture is needed to maintain the reactivity profile of the fuel and thus maintain the necessary reactivity in the full core. While a larger portion of actinides in the resultant MOX fuel will result in greater transmutation, the longer burnup time associated with this fuel results in a higher probability of fuel defects. The

other changing isotopes include Pu-238 (half-life 87.7 years) and Cm-244 (half-life of 18.1 years) which decay over the cooling period by 23.5% and 73.6%, respectively. Finally, Np-237 increases as it is a decay product of Am-241 (half-life of 432 years).

Considering the variations in isotopic content for different cooling periods, there will be differences in lattice cell physics characteristics and in the actinide transmutation over the burnup cycle. Additionally, the actinides remaining at the exit from the burnup cycle will be different affecting the radiotoxicity and longevity of the spent fuel. These issues are explored in the next section.

3. Fuel Composition Variation Due to Aging Studies

Using the aging study composition discussed in Section 2.2, the effects of the different isotopic compositions produced by different cooling times on lattice physics properties, actinide transmutation and end of cycle composition are investigated. Data for the reference TRUMOX-30 fuel transuranic actinide mixture composition is included to provide perspective but the focus of this study is the comparison of the transuranic actinide mixtures from the spent fuel with different cooling times.

3.1 Fresh Fuel Reactivity Effects

Due to the large differences in fissile content and the proportion of tranuranics that have high capture cross sections (e.g., Am-241) there will be a wide variation in the reactivity of the fuel for a consistent blending of the actinide mixture with natural uranium in the TRUMOX-CANDU-900 bundle configuration. Using the reference TRUMOX-30 fuel blending of 3.1% transuranic mixture and 96.9% [7] with the transuranic mixtures of the aging study the variations in fresh fuel reactivity are evident for the different cooling times. These are shown along with the fissile and non-fissile proportions in the transuranic mixture in Table 4.

Table 4 Fresh Fuel Reactivity for Aging Study Fuels (blending 3.1% TRU 96.9%NU)

Property	TRUMOX-30 Reference	5 year cooled	10 year cooled	15 year cooled	20 year cooled	30 year cooled	40 year cooled
Fresh Fuel Reactivity	1.29495	1.35679	1.32521	1.30013	1.28025	1.25238	1.23554
TRU Fissile (wt%)	59.28	60.69	58.53	56.84	55.51	53.67	52.57
TRU Non-fissile (wt%)	40.68	39.31	41.48	43.17	44.49	46.33	47.43

The variation in fresh fuel reactivity is quite wide with reactivity differences of as much as +35 mk for 5 year cooled and as much as -37 mk for the 40 year cooled from the TRUMOX-30 reference value. The change in fresh fuel reactivity is determined as per Equation 1 from the fresh fuel k-infinity values (reactivity) in Table 4.

$$\Delta\rho = \left(\frac{1}{K-inf_{\,reference}} - \frac{1}{K-inf_{\,perturbed}}\right) 1000 \qquad (1)$$

The fissile proportion contributes to the reactivity difference but more significant is the amount of Am-241 present in the fuel. Am-241 has a high capture cross section and will reduce fresh fuel reactivity significantly. For example, the 5 year cooled aging fuel has almost the same fissile content as the reference TRUMOX-30 fuel but has a much higher fresh fuel reactivity because it contains only 3.917 wt% Am-241 compared to 9.907 wt% in the TRUMOX-30 fuel.

The mixing proportions of the transuranic actinides and the NU content will have to be adjusted to maintain the fuel reactivity over the burnup cycle within the operational envelope of the CANDU-900 reactor. For the TRUMOX-30 reference fuel, this corresponds to a fresh fuel reactivity of 1.29495 and an end of cycle reactivity of 0.86446 corresponding to the average burnup of 29,980 MWD/T. This reactivity profile is quite different from the NU fuel used in standard 37 element CANDU fuel which has a fresh fuel k-infinity of 1.12979 and a k-infinity of 0.96680 at the end of its 9,000 MWD/T burnup cycle. The major differences are due to the much higher fissile content of the TRUMOX-30 fuel (2.53 wt% vs. 0.71 wt% for NU) and the presence of the central BNA element in the TRUMOX 43-element bundle design. The differences in the reactivity profile present challenges to the fuelling operations requiring fuelling shifts of 1 to 2 bundles inserted per fuelling visit for the TRUMOX-30 fuel rather than the 4 to 8 bundles inserted per fuelling visit used with natural uranium fuel. The TRUMOX-30 fuel has a longer residency time in the core than natural uranium so the number of fuelling channel visits per day are still close to that of an NU fuelled CANDU unit. Further discussion on the challenges of fuelling TRUMOX-CANDU configurations are found in References [6][7][8][12][13]. The effects on fuelling require full core diffusion modelling and are not explored in this study.

3.2 Burnup Cycle Comparison

The TRUMOX-CANDU envelope is defined using the reactivity profile over the burnup cycle. Using the values from the reference TRUMOX-30 fuel for fresh fuel (1.29495) and end of cycle reactivity (0.86446) the proportions of transuranic actinide mixture from the aging spent fuel and NU can be adjusted to produce a similar reactivity profile over the burnup cycle. The amount of transuranic actinide mix is adjusted such that the fresh fuel lattice cell reactivity is close to the fresh fuel reactivity target set by the reference TRUMOX-30 fuel. The proportions of the TRU mixture and NU for the spent fuel material with different cooling times are provided in Table 5 along with the lattice cell fresh fuel reactivity. These fuel mixtures are used in a TRUMOX, 43-element bundle with the central BNA pin which is simulated within a standard CANDU-900 lattice cell.

Table 5 Aging Study Blending of TRU and NU for TRUMOX-CANDU Reactivity Profile

Property	TRUMOX-30 Reference	5 year cooled	10 year cooled	15 year cooled	20 year cooled	30 year cooled	40 year cooled
Fresh Fuel Reactivity	1.29495	1.29470	1.29495	1.29489	1.29492	1.29493	1.29495
TRU mixture (%)	3.10	1.83	2.32	2.93	3.70	5.83	8.76
NU (%)	96.90	98.17	97.68	97.07	96.30	94.17	91.24

The transuranic feedstock from spent fuel that is cooled for a short time has more Pu-241 and less Am-241 requiring a lower proportion of TRU mixture in the blended fuel to achieve the fresh fuel reactivity target. The range of TRU mixture proportion is 1.83% for 5 year cooled to 8.76% for 40 year cooled. The 40 year cooled fuel has a large amount of Am-241 resulting in high neutron capture in the lattice cell and has the lowest amount of Pu-241 meaning there is less fissile material available. Thus the proportion of TRU mixture needed in the fuel is higher.

The fresh fuel reactivity of the aging fuels is adjusted to match that of the TRUMOX-30 reference fuel and the end of cycle reactivity (0.86446) is used to determine the end of cycle burnup of the fuel. The lattice cell reactivity evolution over the burnup is displayed in Figure 4 for the various aging study fuels and the TRUMOX-30 reference fuel (in the TRUMOX 43-element bundle) and for standard NU-CANDU fuel (37-element, no central BNA, 100% NU).

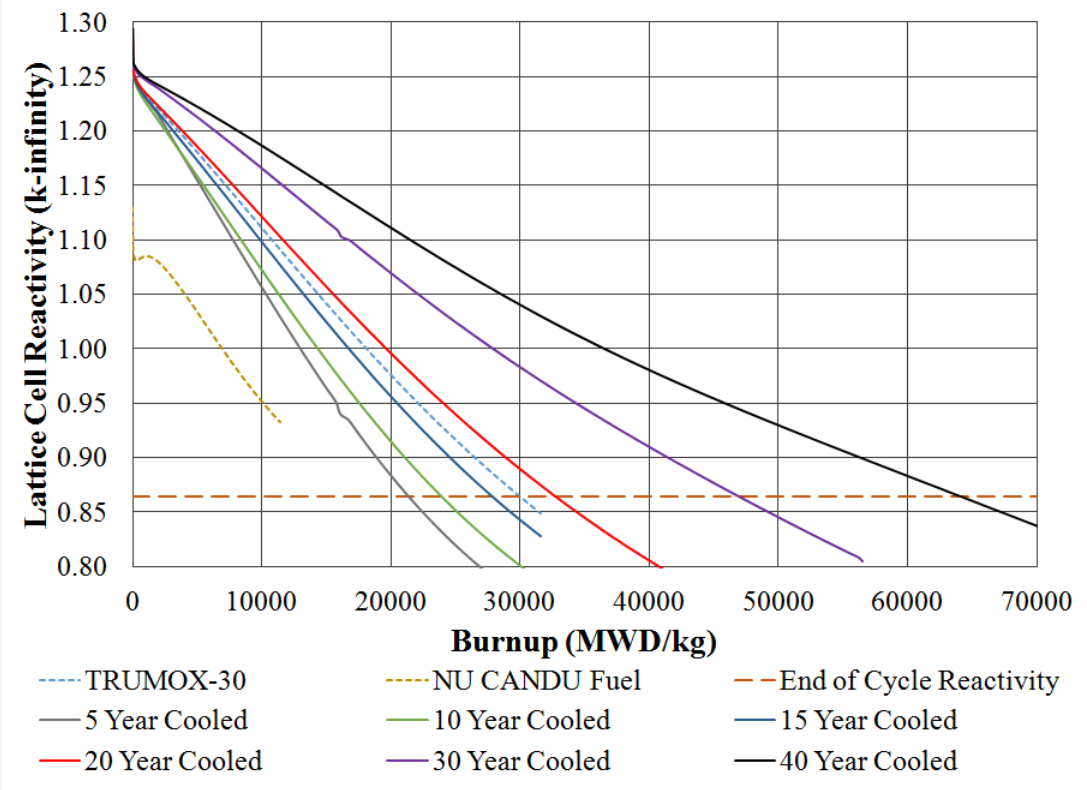

Figure 4 Aging Study Lattice Cell Reactivity Evolution over the Burnup Cycle

The different compositions of the transuranic actinide mixture and the dictated limits on lattice cell reactivity result in different burnup lifetimes of the fuel. As seen in Figure 4, the 15 year and 20 year cooled fuel has a similar slope and burnup lifetime as the TRUMOX-30 fuel. The 5 year and 10 year cooled fuels have a steeper slope and shorter burnup cycle while the 30 year and 40 year cooled fuels have a more gradual slope and longer burnup cycle. When compared to the NU-CANDU fuel, the slope is similar to that of TRUMOX-30 but the reactivity evolution is shifted lower and is much shorter. The dotted line running across Figure 4 is at 0.86446 and

shows the end of cycle burnup for the different fuels. The end of cycle burnups for the aging study fuels are provided along with the TRUMOX-30 fuel in Table 6 along with the transuranic actinide conversion over the burnup cycle.

Table 6 Aging Study Transuranic Actinide Conversion at End of Cycle Burnup

Element Masses (g/bundle)	**TRUMOX-30 Reference**	**5 year cooled**	**10 year cooled**	**15 year cooled**	**20 year cooled**	**30 year cooled**	**40 year cooled**
U-235 initial	119.88	121.44	120.84	120.09	119.14	116.51	112.89
U-235 final	24.54	26.91	25.97	25.08	24.22	22.93	22.51
U-235 change	-95.34	-94.54	-94.87	-95.01	-94.92	-93.58	-90.38
U-235 converted	**-79.53%**	**-77.85%**	**-78.51%**	**-79.12%**	**-79.67%**	**-80.32%**	**-80.06%**
Np initial	24.43	15.57	19.92	25.46	32.59	53.02	82.50
Np final	13.47	9.83	12.09	14.84	18.19	26.99	38.55
Np change	-10.96	-5.74	-7.83	-10.62	-14.40	-26.03	-43.95
Np converted	**-44.87%**	**-36.87%**	**-39.29%**	**-41.70%**	**-44.18%**	**-49.09%**	**-53.28%**
Pu initial	439.23	273.55	338.38	418.99	520.77	802.15	1188.02
Pu final	293.08	205.04	240.64	284.25	338.28	484.08	680.97
Pu change	-146.15	-68.51	-97.74	-134.74	-182.49	-318.06	-507.04
Pu converted	**-33.27%**	**-25.04%**	**-28.89%**	**-32.16%**	**-35.04%**	**-39.65%**	**-42.68%**
Am initial	55.48	16.25	29.13	45.16	65.23	120.31	195.97
Am final	20.87	11.60	16.13	21.73	28.74	48.28	75.95
Am change	-34.61	-4.65	-13.00	-23.43	-36.49	-72.02	-120.02
Am converted	**-62.39%**	**-28.64%**	**-44.63%**	**-51.88%**	**-55.93%**	**-59.87%**	**-61.25%**
Cm initial	0.45	1.34	1.43	1.52	1.62	1.83	2.03
Cm final	11.62	6.94	9.73	12.86	16.44	25.48	37.19
Cm change	11.17	5.60	8.30	11.34	14.82	23.65	35.16
Cm converted	**2491.12%**	**416.39%**	**579.39%**	**746.60%**	**914.61%**	**1292.35%**	**1732.84%**
TRU+Np initial	519.58	306.72	388.86	491.13	620.21	977.30	1468.52
TRU+Np final	339.04	233.41	278.59	333.68	401.67	584.84	832.66
TRU+Np change	-180.55	-73.31	-110.27	-157.44	-218.55	-392.46	-635.85
TRU+Np converted	**-34.75%**	**-23.90%**	**-28.36%**	**-32.06%**	**-35.24%**	**-40.16%**	**-43.30%**
End of Cycle Burnup (MWD/T)	29,980	20,784	23,935	27,845	32,785	46,266	64,035

As expected from the lattice cell reactivity evolution, the 15 year and 20 year cooled fuels have a similar end of cycle burnup as TRUMOX-30 while the 5 year and 10 year cooled fuels have shorter burnup cycles and the 30 year and 40 year cooled fuels have longer cycles. The range of end of cycle burnup is large from 20,784 MWD/T to 64,035 MWD/T. The longer burnup cycles may lead to complications with fuel failures due to stress corrosion cracking and excessive fission gas pressure. Therefore, in order to run fuel for such a long burnup, significant effort and analysis will have to be performed to ensure the fuel can survive with minimal defects. Though

PWR fuels are regularly run to 60,000 MWD/T, regular NU-CANDU fuels are not regularly run much past about 11,000 MWD/T. Previous studies of the TRUMOX-30 fuel provided justification that such a fuel would survive its 30,000 MWD/T burnup cycle with minimal probability of fuel defects [12][13]. Thus the 5 year, 10 year, 15 year and 20 year cooled fuels should have similar performance and survive their predicted burnup cycles with minimal fuel defects. However, the 30 year and 40 year cooled fuels have much longer burnup cycles and may be susceptible to defects. Therefore, the blending of 5 year cooled fuel (more Pu-241 less Am-241) with the older 30 year and 40 year cooled fuel (less Pu-241, more Am-241) transuranic mixtures would serve to produce a blended transuranic mixture that has a shorter burnup cycle around 30,000 MWD/T which would have a lower defect probability.

Based on the determined end of cycle burnup, the transuranic actinide conversion for each of the investigated fuels is computed and the results are provided by element and for overall transuranic conversion in Table 6. In general, the longer the burnup cycle the higher the actinide conversion percentage, additionally the longer burnup fuels in this study require more transuranic mass to meet the reactivity targets resulting in higher overall conversion mass. The issues with long burnup are the higher probability of fuel defects and the significant increases in the production of curium isotopes due to the increased instances of multiple neutron-capture transmuting plutonium and americium to curium.

The conversion percentages achieved for the different fuels vary for the different elements in question. The conversion of neptunium ranges from 36.87% to 53.28% a spread of 16.41%. For plutonium conversion ranges from 25.04% to 42.68% a spread of 17.64%. For americium conversion ranges from 28.64% to 61.25% a spread of 32.61%. For curium, in all cases there is net production ranging from 416.39% to 1,732.84%. This is due to the increased instances of multiple neutron captures transmuting americium to curium as seen in the relevant nuclear reactions shown in Figure 5 [15].

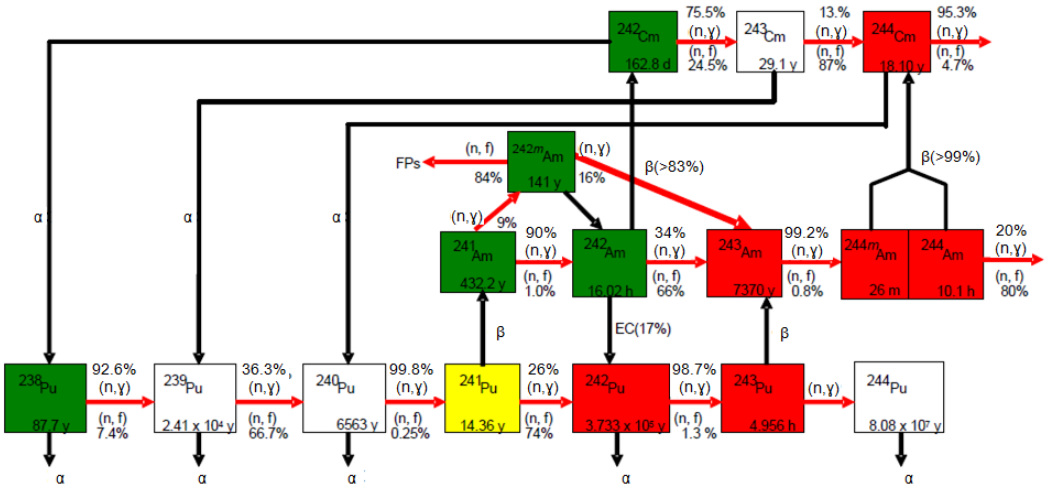

Figure 5 Nuclear Reactions of Interest for Transuranics [15]

Another neutronics property to investigate is the average void reactivity which looks at the lattice cell reactivity (k-infinity) for the standard case (i.e., pressure tube full of coolant) and compares it to the lattice cell reactivity for the case where the coolant in the pressure tube is voided (as can occur in a loss of coolant accident). Coolant void effect is computed by comparing a normal simulation with one where the coolant density is reduced by a factor of $\rho_{liquid} / \rho_{vapour} \approx 1000$. The average void reactivity is measured over the burnup cycle shown in Table 6 for each of the aging study fuels and the TRUMOX-30 fuel, the results are presented in Table 7.

Table 7 Aging Study Transuranic Actinide Conversion at End of Cycle Burnup

Property	TRUMOX-30 Reference	5 year cooled	10 year cooled	15 year cooled	20 year cooled	30 year cooled	40 year cooled
End of Cycle Burnup (MWD/T)	29,980	20,784	23,935	27,845	32,785	46,266	64,035
Average void reactivity (mk)	3.50	0.39	1.66	3.05	4.52	7.60	10.25

The range in average void reactivity for the aging study is 9.86 mk between the low end of 0.39 mk (5 year cooled fuel) and the high end of 10.25 mk (40 year cooled fuel). The TRUMOX-30 reference fuel average void reactivity over the burnup cycle is 3.50 mk [7]. This value is different from a full core void reactivity, which for reference TRUMOX-30 fuel in a CANDU-900 is reported and discussed in Reference [7] as 1.81 mk. This is lower than the full core coolant void reactivity of an NU fueled CANDU of 10-15 mk. While the effects on lattice cell coolant void reactivity provide an idea of the effects of the fuel composition a detailed evaluation of CVR with a full core model is necessary to make comprehensive conclusions.

3.3 Waste Management Consequences

Programs for burning transuranics are already beneficial from a waste management point of view as they remove the long lived actinides from the waste stream leaving only the shorter lived fission products. In the case of burning transuranics in thermal reactors this is to be an intermediary step before moving on to fast reactor systems to compete the transmutation of the transuranics. Considering the aging study explored here, there is a net reduction in all the transuranics in the fuels with the exception of curium. However, there is some production of long lived transuranics over the burnup cycle, especially for longer burnup fuels, which is shown in Table 8 for this aging study.

The longer burnup fuels produce significantly more Pu-242 and Am-243 as well as higher isotopes of curium (Cm-245+) all of which have significantly long half-lives. The curium production issue is less of a problem as there is much less overall mass produced but the 40 year cooled fuel design will produce 6.38 times as much as the 15 year cooled fuel and 4.45 times as much as the 20 year cooled design. This accelerated production becomes a concern for longer burnup fuels (> 20 years). Pu-240 production actually peaks and begins to reverse in fuels that use a transuranic feedstock cooled for 20 years or more this is due to the Pu-240 capturing a neutron and transmuting to Pu-241 which fissions. Eventually, the burning of Pu-240 almost

offsets the production of Pu-242 for 40 year cooled fuel. However, Pu-242 is a much longer lived isotope that captures and produces more Am-243, while Pu-240 has a shorter half-life and transmutes through capture into Pu-241 which is fissionable.

Table 8 Production of Long Lived Transuranics at End of Cycle Burnup

	Mass of isotope produced (g/bundle)						
Isotope (half-life)	**TRUMOX-30 Reference**	**5 year cooled**	**10 year cooled**	**15 year cooled**	**20 year cooled**	**30 year cooled**	**40 year cooled**
Pu-240 (6563 y)	10.79	12.98	9.88	5.53	-0.67	-20.58	-51.38
Pu-242 (3.73×10^5 y)	25.98	17.81	21.08	24.95	29.7	42.00	56.52
Am-243 (7370 y)	7.49	4.58	5.99	7.76	10.00	16.12	23.86
Cm-245 to 248 (> 4730 y)	0.11	0.07	0.11	0.16	0.23	0.52	1.02

Considering the above effects, using older spent fuel feedstock requires longer burnup and thus produces more long lived isotopes of Am, Cm and Pu resulting in increased waste management costs, in both storage and further transmutation of spent MOX fuels. These long lived transuranic isotopes would have to be recycled further in thermal or fast reactors until they are fully transmuted. Considering the half-lives, the spent MOX fuel from a single cycle in a TRUMOX system (i.e. no further reprocessing/recycle) would have a waste longevity similar to that of once through LWR uranium fuel (~100,000 y). To avoid the issues associated with the excessive production of long lived actinides, actinide feedstock from spent fuel with a longer cooling time (30-40 years) could be blended with feedstock from spent fuel with shorter cooling times (< 10 year cooled) to produce a more balanced feedstock that approaches the performance of the 20 year cooled fuel.

3.4 Summary

This aging study has identified possible benefits and drawbacks regarding the use of spent fuel feedstock that has different cooling times. The lattice cell investigations performed provide some insight but are not as rigorous as detailed full core studies looking at fuelling transients, control devices and full core voiding, which would yield more definitive information on fuel performance. Thus this study provides guidance regarding which types of transuranic feedstock may be suitable for further detailed studies.

The TRUMOX-43 element bundle with a central BNA pin used here was originally designed for the TRUMOX-30 reference fuel. If desired, the bundle configuration can be optimized (e.g., BNA loading, burnup cycle time, etc.) for a specific feedstock composition to achieve desired fuel performance. However, this may require more detailed analysis including aspects such as control devices worth, full core void reactivity, power peaking effects of fuelling operations and other characteristics that are best explored with full core analysis.

Using feedstock from spent fuel cooled for short periods of time (< 15 years) presents challenges to fuel manufacture in terms of having high fissile content (specifically Pu-241) which limits the amount of transuranic mixture that can used in the MOX fuel while maintaining within the operational envelope of CANDU-900. The shorter cooled fuels also have low transuranic conversion (< 30%) performance reducing their effectiveness. However, these fuels have low lattice cell void reactivity.

Fuels made from longer cooled transuranic feedstock (> 20 years) have issues with long burnup cycles, which increase the probability of fuel defects (especially > 35,000 MWD/T). The lattice cell void reactivity is larger than the shorter cooled fuels (though still less than standard NU values). Additionally, from a waste management point of view, the use of longer cooled feedstock results in a significantly higher production of long lived transuranics (e.g., Pu-242, Am-243 and higher curium isotopes) which may increase the waste management and follow on irradiation costs for the spent MOX fuels.

Based on the analysis conducted, spent fuels cooled for a medium length of time (~15-20 years) provide a more optimal feedstock balancing the issues of lattice cell reactivity and transuranic conversion rates with those of void reactivity, burnup defects and long term management of the end of cycle waste (i.e., spent MOX fuel).

For the longer cooled feedstock that currently exists, there are options to mitigate the challenges discussed. These include use of separated transuranic extraction (rather than grouped), allowing Am and Cm isotopes to be directed to separate transmutation streams using dedicated targets. Thus providing separate direct transmutation and avoiding including the isotopes in the MOX fuel where they would act as a burnable poison. In regards to challenges with long burnup cycles, it may be useful to explore other reactors which have fuel designs that tolerate longer burnup cycles (e.g., PWR). As mentioned previously, longer and shorter cooled feedstock can be blended together to reach a composition similar to the optimal ~15-20 year cooled feedstock.

4. Conclusion

The variations in isotopic composition for a spent PWR fuel feedstock cooled for different periods from 5 to 40 years was explored for its use in blended mixed oxide fuels. This study focused on group extracted transuranic actinides (Np, Pu, Am, Cm) which are blended with natural uranium to form a MOX fuel. This fuel is then used in a 43-element TRUMOX bundle with a central burnable neutron absorber element in a standard CANDU-900 reactor. Data from the TRUMOX-30 reference fuel composition is included to provide perspective when comparing the MOX fuels derived from PWR spent fuel with different cooling times.

The TRUMOX-CANDU-900 system using the TRUMOX-30 reference fuel composition, has previously undergone detailed reactor physics analysis which has shown it feasible for transuranic actinide burning; achieving significant actinide conversion while maintaining a standard CANDU operational performance envelope [6][7][12][13][14].

MOX fuel cycles, such as presented here, are intended to be used to transmute transuranic actinides in a current thermal reactor design (in this case a CANDU-900 reactor) as an

intermediary step before moving to a fast reactor cycle to complete the transmutation of long lived actinides. This analysis has focused on the CANDU-900 design but previous feasibility studies [8] have indicated that with an appropriate choice of fuel composition (including the transuranic actinide feedstock), this approach is viable with other CANDU designs. Actinide burning is a developing area of research as the removal of the actinides reduces spent fuel radiation and heat loads. The composition of the transuranic actinide feedstock used will affect the lattice cell neutronics of the resulting MOX fuel, the actinide transmutation over the burnup cycle of the fuel and the end of cycle composition which dictates the long term waste management requirements (i.e., further irradiation needs, longevity, radiation and heat loads).

The fuel configurations in this aging study were compared with regards to lattice cell neutronics, transuranic actinide conversion and end of cycle composition. The major change in isotopic composition of spent fuel during cooling is the decay of Pu-241, a fissile isotope, to Am-241, a non-fissile isotope with a high capture cross section, which acts as a burnable neutron absorber in the subsequent MOX fuel. Thus feedstock from fuels with shorter cooling times has higher fissile content and less absorbing isotopes. This limits the amount of transuranic mixture that can be blended with NU to produce a MOX fuel remains within the operational performance envelope of the reactor (defined by lattice cell reactivity profile over the burnup cycle). Fuels with longer cooling times have less fissile material and more absorbing isotopes resulting in lower reactivity and requiring larger proportions of the actinide mixture in the MOX fuel.

The full burnup cycle of MOX fuels composed of NU blended with different transuranic actinide feedstock from spent fuel cooled for different lengths of time was explored through lattice cell calculations using WIMS-AECL. These calculations showed that fuel with shorter cooling times had less actinide transmutation over the burnup cycle and shorter end of cycle burnup reducing the effectiveness of this MOX fueled CANDU system as an intermediary step for actinide transmutation. Use of feedstock from longer cooled spent fuel resulted in MOX fuels with long burnup cycles, increasing the probability of fuel defects. The benefit of significant actinide conversion for these fuels was offset by challenges such as larger lattice cell void reactivity and increased production of long lived transuranics. This presents concerns in terms of neutronics and increased waste management and follow on irradiation costs for the spent MOX fuels. Medium cooled fuel (~15-20 years) was found to strike an acceptable balance of actinide conversion, neutronics performance, burnup cycle and waste management concerns. There is also an option to blend feedstock from spent fuel with short cooling times with that of spent fuel with long cooling times to approximate a composition consistent with that of spent fuel cooled for a medium amount of time which could be mixed with NU to form a suitable MOX fuel.

These conclusions are an estimation based on the spent fuel feedstock explored in this aging study but provide some general insight on the need to balance the proportions of fissionable and non-fissile capture prone isotopes in the transuranic actinide mixture blended into the MOX fuel. The lattice cell analysis performed here illustrates the behaviour of MOX fuels derived from spent fuel that has been cooled for different periods of time but is not exhaustive. Full core analysis taking into account control devices, voiding, fuelling effects are required to explore a chosen fuel in detail.

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