

## The Effects of Fuel Burnup on Incremental Cross Sections in Actinide Fuels in CANDU Reactors

A. C. Morreale<sup>1</sup>, M. R. Ball, D. R. Novog and J. C. Luxat  
McMaster University, Hamilton, Ontario, Canada  
(<sup>1</sup>morreac@mcmaster.ca)

### Abstract

The burning of transuranic actinides from spent fuel in current thermal reactors is an active area of nuclear research. Mixed oxides can be used to combine actinides and natural uranium to produce a high burnup fuel suitable for use in CANDU®\*. Proper safety assessment is needed to ensure standard operational behavior is maintained with advanced fuels. The interaction of control devices with the fuel and the incremental cross sections should be explored over the full burnup cycle of the fuel to determine the changes that may result from composition changes in the fuel over the burnup lifetime.

### 1. Introduction

The reprocessing of transuranic actinides such as neptunium, plutonium, americium and curium from spent fuel, blending them into a mixed oxide (MOX) with uranium and burning them in current reactors is one area of active research helping to close the nuclear fuel cycle [1,2,3]. Studies have been performed of MOX type fuel production and use in current thermal reactor designs, including studies on the feasibility of plutonium disposition in CANDU [4, 5]. Currently PWRs in Europe and Japan operate with a portion of MOX fuel (10-20% of core fuel load). Feasibility studies of a full actinide MOX core have been performed for the CANDU 6 design [6].

The advanced fuels must meet the design requirements for safe operations and reactor control over the full burnup cycle of the fuel. Fuel designs are analyzed for the full core with a neutron diffusion code that derives its inputs from lattice and super-cell calculations. Homogenized cross sections for the fuel, coolant and moderator for a standard cell are provided by the lattice code, while the local reactivity changes caused by interactions between the control devices and the lattice cell are assessed using super-cell calculations. The incremental cross section for a specific control device refers to the differences in the characteristics between the standard lattice cell and the super-cell calculation. This paper studies the effects of fuel burnup on the CANDU incremental cross sections for both natural uranium (NU) and transuranic mixed oxide fuel (TRUMOX) over the burnup cycle.

The two designs explored are the 37 element CANDU bundle fuelled with NU (0.711 wt% U-235) and a 43 element bundle design fuelled with TRUMOX fuel with burnable poison (Dy-Zr) in the central fuel pin. TRUMOX fuel is a blended mixture of actinides extracted from spent LWR fuel (3.1%) and natural uranium (96.9%). The actinide composition is based on yield data from spent fuel reprocessing from Oak Ridge National Laboratories [7]. TRUMOX fuel is designed for longer burnup, 30,000 MWD/T compared to 7500-9000 MWD/T for NU fuel, and as such has a higher fissile content, 2.53 wt% vs 0.71 wt%.

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\*CANadian Deuterium Uranium (CANDU) is a registered trademark of Atomic Energy of Canada Limited (AECL)

## 2. Evaluation of incremental cross sections during burnup

The incremental cross sections for CANDU systems are evaluated using the DRAGON 3D neutron transport code [8]. The CANDU control system places the control devices perpendicular to the fuel channels thus requiring a full 3D super-cell calculation with the device inserted and removed to produce an incremental cross section. The 3-D super-cell contains two fuel channels with a control device in between the channels, see Figure 1.

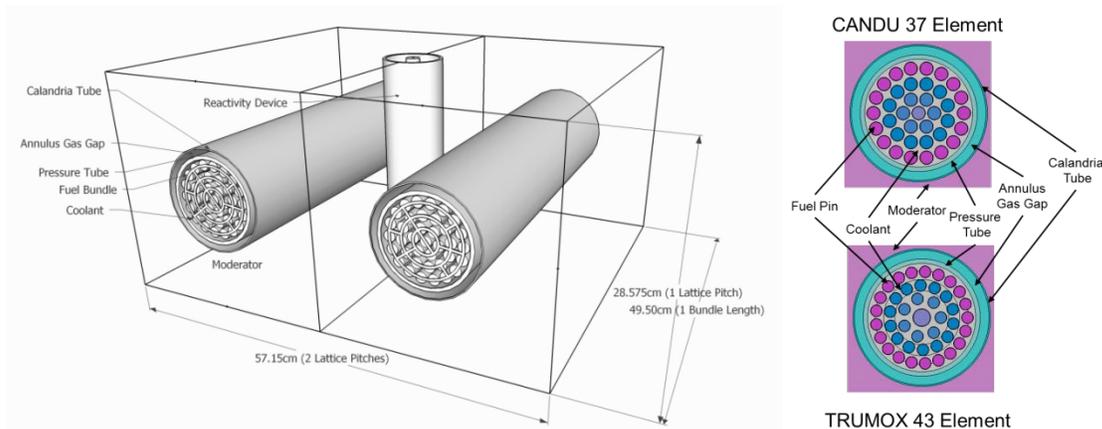


Figure 1 & 2 DRAGON 3D Super-cell Model(37 element)and 2D lattice cell models.

The 3D super-cell model run in DRAGON uses a fully detailed 2D model detailing the 37 or 43 elements of the fuel bundle for the self-shielding calculation, but the 3D transport solution is performed with annularized bundle properties. Specifically, the outer rings (rings 2, 3, 4) are depicted as homogenized annuli of fuel, coolant and cladding material rather than individually clad elements in coolant. The annular fuel rings maintain the volume of fuel, coolant and cladding to represent the fuel elements at the common radial distance from the centre of the bundle.

A 2D infinite lattice simulation of the cluster model is used for the two fuel types, see Figure 2. The NU bundle has 37 elements with a common diameter arranged in concentric rings of 1, 6, 12, and 18 elements while the TRUMOX bundle has a large center element surrounded by concentric rings of 7, 14 and 21 smaller elements. The 2D simulations are performed for the full burnup cycle of each fuel and the fuel composition in each ring is extracted for the desired burnup level and is used as an input into the 3D super-cell calculation. A power level of 32 W/g and exit burnups of 9000 MWD/T for NU and 30,000 MWD/T for TRUMOX are used.

The incremental cross section simulations are performed with a stainless steel (pin in tube) adjuster rod interacting with the system of the 37 element NU bundle and the 43 element TRUMOX bundle. The control device is modelled at 0% and 100% inserted and the 2 group homogenized cross sections are computed. The fast and thermal incremental cross sections:  $\Delta\Sigma_{\text{Total}}$ ,  $\Delta\Sigma_{\text{absorption}}$ ,  $\Delta\Sigma_{\text{N-Gamma}}$ , and  $\Delta\nu\Sigma_f$  along with the  $\Delta\Sigma_{\text{scatter1, 2}}$  and  $\Delta\Sigma_{\text{scatter2, 1}}$  are computed. The incremental cross section at several specific burnup levels is analysed for the two fuel types. In general, incremental cross section calculations are performed at mid-cycle burnups this most closely approximates the fuel composition that the devices will interact with during reactor operations.

### 3. Simulations and Results

Both the NU and TRUMOX fuels undergo composition changes during their burnup cycle that have effects on the neutronics and cross section behaviour of the fuel, Figure 3. The U-238 in the NU fuel undergoes neutron capture producing Pu-239 adding fissile content to the depleting amount of U-235. The fissile content in NU is dominated by U-235 during most of the cycle but towards the end the Pu-239 content surpasses that of U-235 (~6800MWD/T). For TRUMOX, the fissile content is dominated by Pu-239 for the full burnup and the concentrations of all fissile elements (Pu-239, Pu-241 and U-235) constantly decline over the burnup cycle.

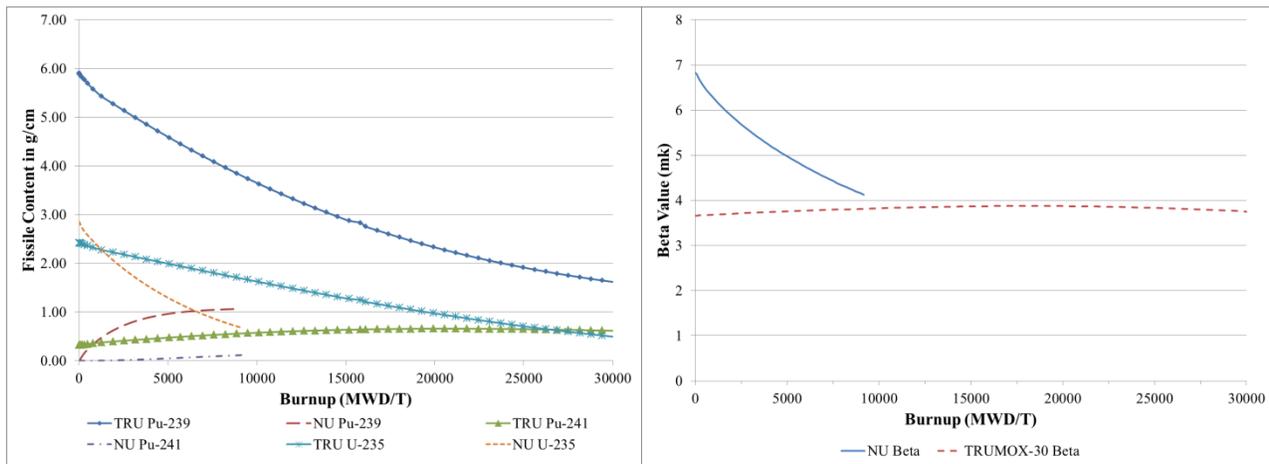


Figure 2& 4fissile isotope concentrationsand delayed neutron fraction ( $\beta$ ) over the burnup cycle

The changing fuel composition alters the delayed neutron fraction,  $\beta$ , affecting the relative dollar worth of control devices. The single lattice cell  $\beta$  is tracked over the relevant burnup cycle for each fuel based on the composition of fissile isotopes in Figure 4. It should be noted that a full core with many channels all various burnups will produce an average  $\beta$  for the full core based on the overall fuel composition (NU CANDU  $\beta_{Full\ Core} = 5.82\text{mk}$ ) [10]. The NU beta value is initially high (6.82mk) and decreases as plutonium is produced in the fuel (down to 4.15mk at 9000 MWD/T, a drop of 2.67mk over the burnup cycle). The TRUMOX fuel has higher plutonium content and thus lower  $\beta$  values (~3 mk less than NU) that start at 3.66 mk and rise to a peak of 3.88 mk and then decrease to 3.75 mk at 30000 MWD/T (a range of 0.22 mk), which is more stable than the NU case.

The lattice cell reactivity changes experiences similar changes over the burnup for both fuel types, an initial strong reduction followed by a linear decline as fission content is steadily depleted. The initial decrease results from the production of neutron absorbing fission products, such as Xe-135, which build-up to a steady state value in the first week of operations. In NU, the U-238 transmutes to Pu-239 adding fissile content to the fuel. Early in the cycle these gains outpace the losses due to burnup producing a reactivity peak at about 1100MWD/T. The TRUMOX fuel has a higher fissile content, mostly Pu, and a reduced U-238 content which makes the transmutation to Pu-239 a less dominating effect on the fuel reactivity and hence there is no reactivity peak. Compared to NU fuel, TRUMOX is more active and hence its initial k-infinity is 12.6% higher. It is also 10.7% higher at the NU reactivity peak (1120MWD/T) but falls below that of NU (9.9% lower) at the end of the

TRUMOX burnup cycle. For a critical operating reactor with fuels of various burnups, active new fuel balances out older fuel. Since TRUMOX is more active to begin with it is able to stay in core longer reaching a lower k-infinity value while maintaining full core reactivity.

Table 3: Effects of Burnup on Super-cell Reactivity Change from Adjuster Rod Insertion

Natural Uranium Fuel			TRUMOX Fuel		
Burnup(MWD/T)	$\Delta\rho$ (mk)	$\Delta\rho/\beta$ (\$)	Burnup(MWD/T)	$\Delta\rho$ (mk)	$\Delta\rho/\beta$ (\$)
0	52.7	7.23	0	26.8	7.33
1760	50.1	8.41	4480	29.8	7.96
3680	49.9	9.36	9120	32.6	8.54
4480	50.2	9.81	13600	35.5	9.21
7520	51.9	11.71	20640	40.4	10.44
9120	52.9	12.77	27680	45.3	11.93

There are differences in the reactivity changes for adjuster insertion between the NU and TRUMOX fuels over their burnup cycles as shown in Table 1 and Figure 5. The reactivity change for NU decreases initially and then after a flat period begins to linearly increase as the middle of the burnup cycle is reached (total range = 3 mk). The initial decline in NU is caused by plutonium build-up in the fuel during the first half of the cycle. The TRUMOX fuel undergoes a linear increase in reactivity change throughout the cycle that increases from 26.8 mk to 45.3mk, a range of 18.5 mk.

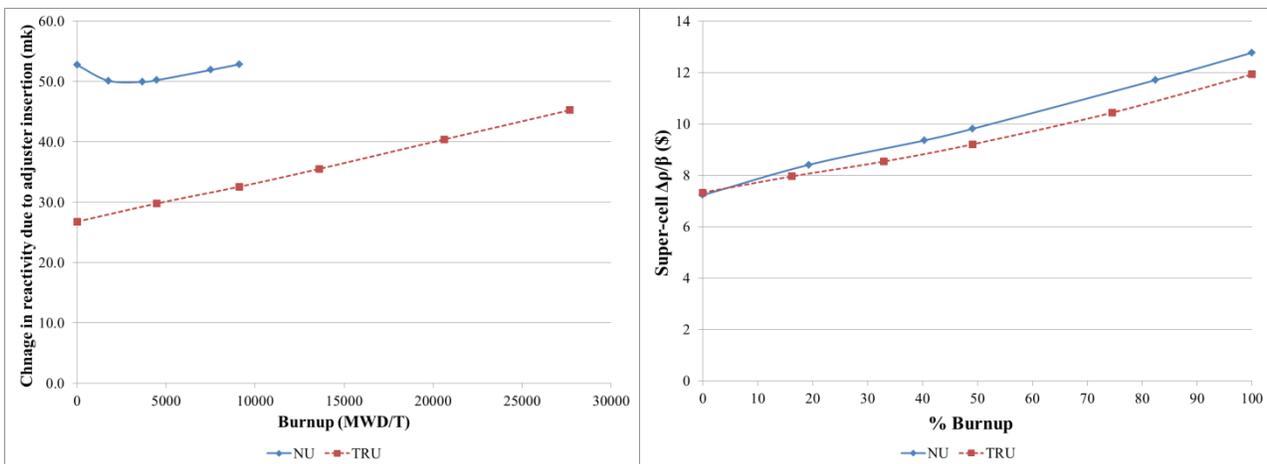


Figure 5 & 6 Effects of burnup on absolute and relative ( $\Delta\rho/\beta$ ) reactivity change due to rod insertion

Using the  $\beta$  values computed in Figure 4, the relative reactivity change, ( $\Delta\rho/\beta$ ), was computed for each of the burnup steps. The two fuels have similar linear increasing relative reactivity when compared over their relative burnup cycles as seen in Figure 6. The dollar values for TRUMOX are 1.38% higher than NU for fresh fuel, 6.12% lower at the mid burnup stage and 6.58% lower at the end of burnup. Despite the different burnup behaviour of the two fuel types, their relative reactivity performance is quite comparable over their burnup cycle suggesting similar controllability limits. This is an important issue when considering the use of TRUMOX fuel in an NU CANDU system.

### 3.1 Effects of burnup on incremental cross sections

The effects of burnup on fuel behaviour are also seen in the changes in the incremental cross sections of the fuels for the adjuster rod insertion. The incremental cross sections examined in these trials include fast and thermal  $\Delta\Sigma_{\text{Total}}$ ,  $\Delta\Sigma_{\text{absorption}}$ ,  $\Delta\Sigma_{\text{N-Gamma}}$ , and  $\Delta v\Sigma_f$  along with the  $\Delta\Sigma_{\text{scatter1, 2}}$  and  $\Delta\Sigma_{\text{scatter2, 1}}$ . These were computed for several burnup steps for each of the fuel types, see Table 2.

Table 2: Effects of Burnup on Incremental Cross Sections ( $\Delta\Sigma$ ), NU Fuel and TRU Fuel

	Burnup (MWD/T)	GR 1 (Fast) NU-SIGMA-F	GROUP 2 (Thermal)				Up Scatter Gr2 to Gr1
			Total	Absorption	N-Gamma	NU-SIGMA-F	
NU Fuel $\Delta\Sigma$ (cm <sup>-1</sup> )	0	-2.12E-07	2.31E-04	2.45E-04	2.42E-04	7.70E-06	4.64E-06
	1760	-2.26E-07	2.47E-04	2.51E-04	2.45E-04	1.49E-05	4.72E-06
	3680	-2.39E-07	2.54E-04	2.53E-04	2.47E-04	1.80E-05	4.72E-06
	4480	-2.43E-07	2.56E-04	2.54E-04	2.47E-04	1.87E-05	4.72E-06
	7520	-2.52E-07	2.60E-04	2.55E-04	2.48E-04	1.99E-05	4.70E-06
	9120	-2.54E-07	2.62E-04	2.55E-04	2.48E-04	2.01E-05	4.69E-06
	<b>% Change</b>	19.77%	13.12%	4.15%	2.56%	161.18%	1.08%
TRU Fuel $\Delta\Sigma$ (cm <sup>-1</sup> )	0	-2.24E-07	3.36E-04	2.78E-04	2.66E-04	3.43E-05	4.67E-06
	4480	-2.21E-07	3.31E-04	2.76E-04	2.65E-04	3.21E-05	4.66E-06
	9120	-2.29E-07	3.24E-04	2.74E-04	2.63E-04	2.99E-05	4.64E-06
	13600	-2.37E-07	3.18E-04	2.72E-04	2.62E-04	2.79E-05	4.62E-06
	20640	-2.47E-07	3.09E-04	2.69E-04	2.60E-04	2.52E-05	4.59E-06
	27680	-2.53E-07	3.01E-04	2.66E-04	2.58E-04	2.32E-05	4.57E-06
	<b>% Change</b>	13.04%	-10.20%	-4.16%	-2.87%	-32.32%	-2.26%

For both fuels, there is very little change over the burnup for the group 1 (fast)  $\Sigma_{\text{Total}}$ ,  $\Sigma_{\text{absorption}}$ ,  $\Sigma_{\text{N-Gamma}}$  and the  $\Sigma_{\text{scatter1, 2}}$  values (less than  $\pm 1.0\%$ ). The group 1  $v\Sigma_f$  values both increase with the NU case increasing by 6.8% more than the TRUMOX case. In group 2 (thermal), the two fuel types have opposite responses, the changes are all positive in the NU fuel case and all negative in the TRUMOX case. The behavior over the burnup for the two fuels is very different for the thermal  $\Sigma_{\text{Total}}$ ,  $\Sigma_{\text{absorption}}$ ,  $\Sigma_{\text{N-Gamma}}$  (Figure 7) and  $v\Sigma_f$  values. In the NU case the incremental values generally increase sharply at the beginning and plateau during the last half of the burnup while the TRUMOX values decrease linearly with burnup. These differences are caused by the plutonium build up in the NU fuel described earlier as the incremental cross sections to follow the Pu-239 concentration during burnup. In the specific case of thermal absorption in TRUMOX, the decrease over burnup is driven by actinides with high thermal absorptions that have been transmuted to other isotopes with lower absorption cross sections. Over the full burnup cycle about 35% of the higher actinides, many of which have high thermal absorption cross sections will be transmuted.

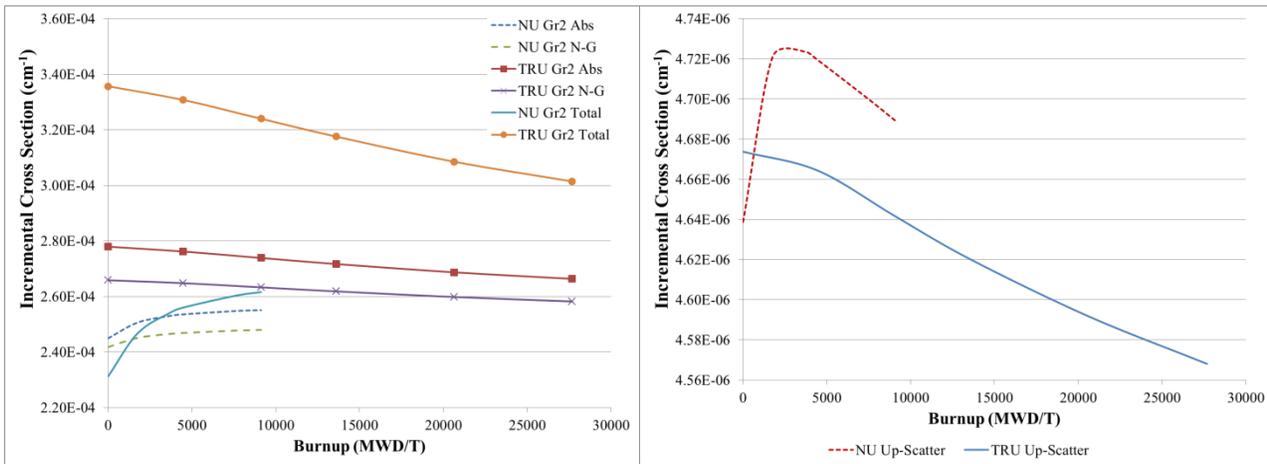


Figure 7&8 Change in thermal and Up-scatter incremental cross sections over the burnup cycle

For the up-scatter incremental cross section, Figure 8, the NU case increases sharply to a maximum in the early burnup, plateaus shortly and then decreases linearly for the last half of the burnup. The TRUMOX case maintains a steady decline that is fairly linear. The percent change in up-scatter is proper for the TRUMOX case, a loss of 2.26%, but is inaccurate for NU as the change is -0.68% if measured from the peak rather than 1.08% as measured from the fresh fuel initial value.

Comparison between the NU and TRUMOX cases can also be made at relative places in the burnup cycle (ex. Fresh, mid, end). Table 3 provides the percent difference of the TRUMOX incremental values from those of NU. The group 1 (fast) values for  $\Sigma_{Total}$ ,  $\Sigma_{absorption}$ , and  $\Sigma_{N-Gamma}$  are relatively small (all within  $\pm 1.2\%$ ) as are the values for scattering.

Table 3: Comparison of NU and TRUMOX Incremental Cross Sections at Burnup Stage

Burnup Stage	Gr 1 (Fast) NU-SIGMA-F	Gr 2 (Thermal) – % diff from NU(TRU-NU)/NU				Up Scatter Gr2 to Gr1
		Total	Absorption	N-Gamma	NU-SIGMA-F	
Fresh Fuel	5.57%	45.15%	13.46%	9.94%	345.20%	0.76%
Mid Burnup	-2.44%	24.05%	7.15%	6.06%	49.31%	-2.10%
End of Burnup	-0.37%	15.23%	4.41%	4.12%	15.37%	-2.58%

\* Mid Burnup: NU = 4480 MWD/T, NU End of Burnup = 9520 MWD/T  
 \*\* TRU Mid Burnup = 13600 MWD/T, TRU End of Burnup = 27680 MWD/T

The group 1  $v\Sigma_f$  value is above NU for fresh fuel but drops below as burnup increases since the reactivity of the TRUMOX fuel is lower than NU at the respective end of burnup. The up-scatter differences range from +0.76% down to -2.6%. The biggest differences from NU are in the group 2 (thermal) values for  $\Delta\Sigma_{Total}$  and  $\Delta v\Sigma_f$ . The total values are 45% higher for fresh fuel and decrease to 15% over the burnup cycle. The  $\Delta v\Sigma_f$  is very different for fresh fuel (345%) due to the much higher fissile content of the TRUMOX fuel (~ 3.5 times that of NU, 2.53% vs 0.71%). As the fuel burns, the fissile content decreases and by the time both fuels reach the end of their burnup cycle the difference is about 15% since the TRUMOX burnup cycle length that is 3 times as long as NU.

#### 4. Conclusion

The analysis of the effects of fuel burnup on the adjuster incremental cross sections in a CANDU reactor for both NU and TRUMOX fuel was performed using DRAGON with 2D burnup and 3D incremental simulations. The use of higher enriched fuels such as TRUMOX with higher plutonium content and harder neutron spectra reduce the reactivity change during adjuster insertion due to the lower capture cross section of stainless steel and also affect the adjuster incremental cross sections. The NU fuel experiences a reactivity peak around 1,100MWD/T due to the transmutation of U-238 to Pu-239 which affects the reactivity change and adjuster incremental cross section response. This peak is not present in the TRUMOX fuel as it already has high plutonium content and lower U-238 concentration. Despite the different composition and delayed neutron factor behaviour during the burnup cycles the relative reactivity changes due to rod insertion for the two fuels are similar indicating similar controllability. The adjuster incremental cross sections tend to rise to a plateau for the NU case over the burnup cycle with the rise consistent with the period before the plutonium peak while the TRUMOX case shows a constant linear decline during the burnup cycle. The biggest effects are in the adjuster incremental cross sections for thermal fission and are related to the amount of fissile material in the fuel. This information can be utilized as input to full core diffusion calculations of a TRUMOX fuelled reactor to provide more realistic simulations suitable for detailed analysis and the modelling of accident scenarios.

#### 5. References

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