Material Balance of Fuel Cycles for Plutonium Recycling in Pressurised Water Reactors

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Abstract. An inevitable effect of uranium fuel operation is the production of plutonium. Its increasing inventory requires adoption of plutonium management strategy. The option that can provide inventory minimization and utilization of plutonium's energy content is its usage as nuclear fuel. Since the most common power reactors are the pressurized water reactors, the current paper examines the material balance of several fuel cycles for single and multiple plutonium recycle in a reference PWR. Once-through fuel cycle's balance has been used as a benchmark. Plutonium production and consumption rates, uranium and separative work requirements have been evaluated for four fuel cycle variations. The results affirm the overall long-term feasibility of multiple plutonium recycle in PWRs in terms of increased plutonium consumption.

1 Introduction

An inevitable effect of uranium fuel operation is the production of plutonium. Its increasing inventory requires adoption of plutonium management strategy. The annually generated plutonium in power reactors varies between 136 and 366 kg per reactor, depending on the reactor type [1]. The latest estimation, dating from 2015, shows that at the end of 2014 the global civilian plutonium stockpile amounted at 271 t HM [2]. Generally, there are three approaches for plutonium management – indefinite storage in facilities with high levels of physical protection; manufacturing of mixed uranium-plutonium fuels and burning in commercial reactors; immobilization in glass or ceramic matrix and final disposal [3]. The option that can provide inventory minimization and utilization of plutonium's energy content is its usage as nuclear fuel.

Mixed uranium-plutonium oxide fuels (MOX), used in light water reactors, usually contain 4-10 wt. % of plutonium [4]. Most often, for MOX fuel manufacturing depleted uranium is used. That allows increasing the plutonium concentration in the fuel due to the low fraction of fissile material in the depleted uranium [5].

A variation of mixed uranium-plutonium fuel that uses enriched, instead of depleted uranium, is known as MOXEUS (**MOX E**nriched **U**ranium **S**upport), MOX-EU (MOX-**E**nriched **U**ranium) or MIX. It is suitable for multiple plutonium recycle in pressurised water reactors. In this fuel the main source of fissile material is the enriched uranium. Since

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plutonium has only a secondary role, the fuel characteristics are similar to those of uranium oxide (UOX) fuel. The enriched uranium is used to compensate for the reactivity loss due to the deteriorating plutonium isotopic composition that is observed in the case of multiple irradiations. That allows multiple plutonium recycling in pressurised water reactors [4,6].

2 Objective and input data

The main objective is to carry out a comparative analysis of four fuel cycle options – a once trough cycle and three closed cycles in order to determine the plutonium mass balance and to outline the best option in terms of plutonium inventory management. The assessed fuel cycle options are presented on Figure 1. The analysis includes determination of the fuel cycles' material balances. The needed calculations have been carried out using the code ORIGEN, part of the SCALE Version 6.1 software package [7].

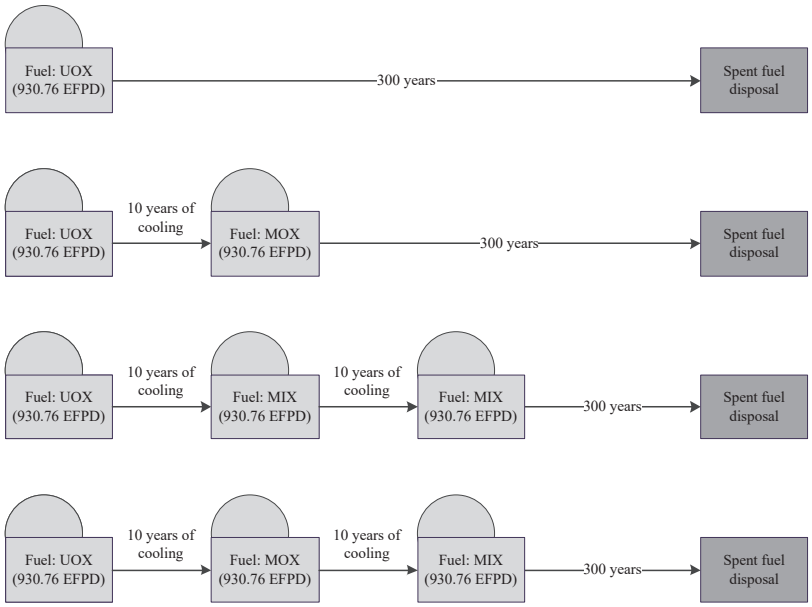

Fig. 1. Schematic representation of the analysed fuel cycles. From top to bottom: case 1 to case 4.

In all four cases a reference pressurised water reactor (PWR) with installed capacity of 1000 MW and gross thermodynamic efficiency of the power unit of 33% has been used in the calculations. In the once-through cycle the load is 4.8% enriched uranium oxide fuel (UOX) with design burn-up of 62,000 MWd/tHM. In the case of closed fuel cycles, the spent fuel of each stage is cooled down for 10 years after discharge before reprocessing, extracting the plutonium and mixed oxide fuel manufacturing. Two options for mixed uranium-plutonium fuels have been chosen: the traditional MOX fuel and the MIX concept. In order to achieve comparability, the mixed fuels have burn-up of 62,000 MWd/tHM. The analysed fuel cycle cases are as follows:

1)Case 1: once-through cycle; the spent fuel is cooled down for 300 years before disposal.
2)Case 2: single recycle of plutonium in the form of MOX fuel and subsequent cool down of 300 years;
3)Case 3: double plutonium recycle in the form of MIX fuel with intermediate 10-year cooling periods between stages and final 300-year cool down before disposal;

4)Case 4: double plutonium recycle; the first stage uses MOX and the second – MIX fuel, with intermediate 10-year cooling periods and final 300-year cool down of the spent MIX fuel before disposal.

The input data for the calculations are summarised in Table 1.

Table 1. Input data for the analysis.

Indicator	Unit	UOX	MOX	MIX
Installed electric capacity	*MW*	1,000.00	1,000.00	1,000.00
Gross thermodynamic efficiency	*%*	33.00	33.00	33.00
Fuel burn-up	*MWd/tHM*	62,000.00	62,000.00	62,000.00
Capacity factor	*%*	85.00	85.00	85.00
Enrichment	*wt.%*	4.80	-	4.80
^{235}U weight fraction in depleted uranium	*wt.%*	-	0.30	-
Plutonium weight fraction	*wt.%*	-	7.00	7.00
Fuel cycle length	*years*	3.00	3.00	3.00
Fuel cycle length	*EFPD*	930.75	930.75	930.75
Total mass of fuel loaded	*tHM*	45.49	45.49	45.49
Enriched uranium mass	*tHM*	45.49	-	42.31
Mass of plutonium loaded	*tHM*	-	3.18	3.18
Mass of depleted uranium used	*tHM*	-	42.31	-
Thermal power	*MW*	3,030.30	3,030.30	3,030.30
Gross power generation	*TWh*	22.338	22.338	22.338
Average thermal power	*MW/tHM*	66.61	66.61	66.61

Enrichment, burn-up, plutonium weight fraction and fuel cycle lengths have been chosen based on [6,8]. The total mass of fuel loaded G_x, the thermal power of the reactor P, the effective fuel cycle length $T_{eff.}$, expressed in effective full-power days (EFPD), and the gross electric output $W_{gr.}$, have been calculated using Eq. (1) [9]:

$$G_x = \frac{Q}{B} = \frac{W_{gr.}}{B \cdot \eta} = \frac{N \cdot T_{eff.}}{B \cdot \eta} = \frac{N \cdot \varphi \cdot T_c}{B \cdot \eta} \quad (1)$$

In Eq. (1) the designations are as follows: N – electric capacity of the nuclear power unit, MW; □ – gross thermodynamic efficiency of the nuclear power unit; B – nuclear fuel burn-up, MWd/tHM; □ – capacity factor; T_c –calendar length of the fuel cycle, days. The average thermal power q_m has been calculated using Eq. (2):

$$q_m = \frac{P}{G_x} \tag{2}$$

The data is used as an input in ORIGEN to calculate the concentrations of plutonium isotopes in the spent fuel for each fuel cycle. These results are shown in Table 2. The obtained isotopic vectors are used to determine the isotopic compositions of the fresh fuels (Table 3).

Table 2. Isotopic vectors of the plutonium, used for mixed fuel manufacturing, wt. %.

Nuclide	MOX after UOX	MIX after UOX	MIX after MOX)	MIX after MIX
^{238}Pu	3.57%	3.57%	3.62%	4.01%
^{239}Pu	51.17%	51.17%	35.81%	36.71%
^{240}Pu	25.76%	25.76%	31.26%	32.00%
^{241}Pu	9.93%	9.93%	11.75%	11.53%
^{242}Pu	9.56%	9.56%	17.56%	15.75%

Table 3. Fresh mixed fuels' isotopic compositions, wt. %.

Nuclide	MOX after UOX	MIX after UOX	MIX after MOX	MIX after MIX
^{238}Pu	0.25%	0.25%	0.25%	0.28%
^{239}Pu	3.58%	3.58%	2.51%	2.57%
^{240}Pu	1.80%	1.80%	2.19%	2.24%
^{241}Pu	0.70%	0.70%	0.82%	0.81%
^{242}Pu	0.67%	0.67%	1.23%	1.10%
^{235}U	92.72%	88.54%	88.54%	88.54%
^{238}U	0.28%	4.46%	4.46%	4.46%

3 Results and analysis

The change over time of the total concentration of plutonium isotopes is presented on Figure 2. The concentration is the lowest in the case of once-through cycle because plutonium is produced as a result of a neutron capture by ^{238}U nuclei. At discharge, the plutonium concentration is 13 kg/tHM and increases to 13.169 kg/tHM after 0.3 years. That

can be explained by the growing concentration of ^{238}Pu, ^{239}Pu and ^{240}Pu due to the decay of heavier short-lived transuranic nuclei. In the period between 0.3 and 300.0 year, the amount of plutonium in the spent fuel slowly decreases to 10.816 kg/tHM. The greatest quantity of plutonium is generated in the UOX-MOX cycle (case 2). At discharge it is 47.12 kg/tHM, decreasing to 38.764 kg/tHM at year 300.0. This can be explained with the high initial plutonium weight fraction in the fresh MOX fuel – 7 wt. %, and the rest being mainly ^{238}U that contributes to further plutonium generation.

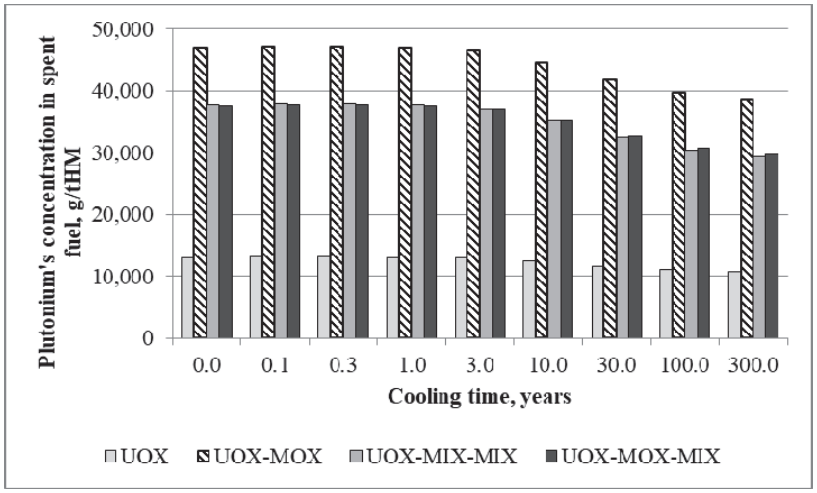

Fig. 2. Plutonium concentration in spent fuel to be disposed of in different fuel cycles as a function of cooling time.

From figure 2 it can be seen that in cases 3 and 4 (UOX-MIX-MIX and UOX-MOX-MIX) the plutonium concentration is similar over time. In the same time, it is lower than in the single recycle case. That can be explained with the accumulation of heavier minor actinides that are produced by neutron capture in plutonium nuclei. That is due to the generally higher concentrations of heavier plutonium isotopes in MIX fuel. Moreover, the concentration of ^{238}U in MIX fuel is lower.

The summarised values of the indicators of the material balance are outlined in Table 4. The values are calculated for each of the fuel types used in the analysed fuel cycle cases. In all closed fuel cycle cases a negative growth rate of the plutonium is observed. The highest plutonium consumption is in the case of double recycle using MIX, manufactured from spent MOX fuel (case 4). The consumption in this occasion is with 0.33 kg Pu/TWh higher than in the other considered option for double recycle.

The results show an advantage of the closed fuel cycles in terms of diminished separative work requirements: in the case of MIX fuel manufacturing, it is about 1 tSW/TWh lower compared to uranium fuel manufacturing. In the case of MOX fuel manufacturing there are no separative work requirements. In terms of depleted uranium consumption, such is observed only in the case of MOX fuel manufacturing because of the initial composition of this fuel type – plutonium and depleted uranium.

Figure 3 schematically represents the normalized plutonium growth rate (generated plutonium per terawatt-hour of gross electricity production) at each stage of the analysed fuel cycle cases. The only case in which a positive plutonium growth rate is observed is case 1 (once-through cycle). The growth rate is 26.57 kg/TWh or about 600 kg present in the spent fuel.

Table 4. Material flows at each fuel cycle stage.

Indicator/Stage	UOX	MOX	MIX after UOX	MIX after MIX	MIX after MOX
Total fuel load, tHM	45.49	45.49	45.49	45.49	45.49
Enriched uranium mass, tHM	45.49	-	42.31	42.31	42.31
Mass of plutonium loaded, tHM	-	3.18	3.18	3.18	3.18
Mass of depleted uranium loaded, tHM	-	42.31	-	-	-
Enriched uranium consumption, tHM/TWh	2.04	-	1.89	1.89	1.89
Natural uranium consumption, tHM/TWh	22.30	-	20.74	20.74	20.74
Depleted uranium stockpile, tHM/TWh	20.26	-1.89	18.84	18.84	18.84
Uranium concentrate consumption, t/TWh	26.30	-	24.45	24.45	24.45
Separative work, SWU	309,776.44	-	288,099.59	288,099.59	288,099.59
Normalized separative work, tSW/TWh	13.87	-	12.90	12.90	12.90
Plutonium mass at fuel loading, kg HM	0.00	3,184.30	3,184.30	3,184.30	3,184.30
Plutonium mass at fuel discharge, kg HM	591.37	2,143.49	2,312.71	1,718.16	1,710.88
Plutonium growth rate for the cycle, kg HM	591.37	-1,040.81	-871.59	-1,466.14	-1,473.42
Normalized plutonium growth rate, kg HM/TWh	26.47	-46.59	-39.02	-65.63	-65.96

In all closed fuel cycles plutonium consumption is observed because the burning rate due to fission and neutron capture is higher than the production rate due to secondary fuel breeding. After the first plutonium recycling step using MOX (cases 2 and 4) the growth rate is -46.59 kg/TWh. That amounts to a little more than 1 t of burnt plutonium. The recycling stage using MIX consumes 39.02 kg/TWh plutonium. The difference of 7.57 kg/TWh is due to the fact that the main fissile isotope in MOX is ^{239}Pu, as opposed to MIX fuel where the main fissile material is ^{235}U.

In the case of double recycle using MIX the plutonium consumption rate is about 66 kg/TWh, which is increase of around 40% compared to MOX. That increase might be

explained with neutron capture by even plutonium isotopes whose concentrations increase in multiple recycle cases.

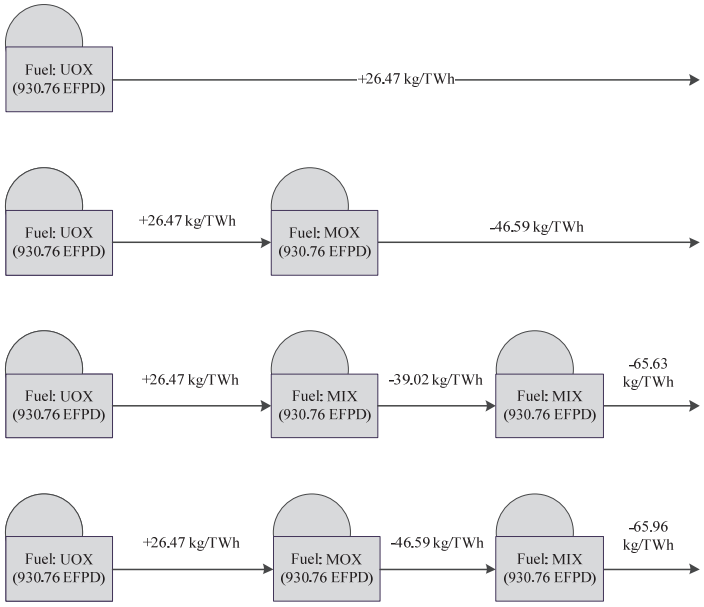

Fig. 3. Plutonium material flows at each stage of the analysed fuel cycles.

The summarised results of the plutonium growth rate for each fuel cycle case are outlined in Table 5. The different electricity production in each case is taken into account. It can be concluded that, in terms of plutonium inventory management, the most efficient is case 4 – multiple recycle using MOX fuel in the first recycle stage and MIX fuel in the second recycle stage.

Table 5. Normalized plutonium growth rate.

Fuel cycle case	Gross power generation in the fuel cycle, TWh	Normalized plutonium growth rate of the fuel cycle, kg HM/TWh
UOX (Case 1)	22.338	26.47
UOX-MOX (Case 2)	44.676	-10.06
UOX-MIX-MIX (Case 3)	67.014	-26.06
UOX-MOX-MIX (Case 4)	67.014	-28.69

5 Conclusions

Plutonium recycling leads to savings of natural uranium – the primary nuclear fuel resource. Core loading with mixed uranium-plutonium fuels leads also to uranium concentrate, conversion, and separative work savings. In the case of 100% MOX loading natural uranium savings amount to 100% since depleted uranium is used for mixed fuel manufacturing.

In the cases of multiple plutonium recycles using MIX, those savings are lower due to enriched uranium usage and decreasing concentration of fissile plutonium isotopes with each recycle step.

In terms of plutonium growth rate, it is positive only in the once-through cycle. The absolute value of the growth rate in the closed cycles depends on the isotopic composition of the fresh fuel and the irradiation regimes. At the first recycle stage, MOX fuel usage results in higher plutonium consumption, compared to MIX fuel. That is a consequence of plutonium being the main fissile material in MOX fuel, as opposed to MIX, where the main fissile isotope is ^{235}U. In terms of overall consumption, however, the most efficient fuel cycle is case 4: multiple recycle using MOX fuel in the first recycle stage and MIX fuel in the second recycle stage. That is due to increased consumption of plutonium in the second recycle stage.

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