PROLIFERATION RESISTANCE OF NUCLEAR FUELS FOR SMALL MODULAR REACTORS

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Summary

Proliferation resistance has become one of the primary topics to be addressed if new nuclear energy systems are being developed as any nuclear system presents potential proliferation risks. In a near future, the diffusion of a nuclear fleet of Small Modular Reactors (SMRs), with inherent safety characteristics and with low level of attractiveness to the proliferation, could further enhance the capability to utilize plutonium from spent fuel and weapons dismantling, so providing a new option for reducing civil and military plutonium stocks. In this context, the objective of the present study is to identify the level of proliferation attractiveness of fuel loaded in SMR cores based on pressurized light water technology (PWR). In order to face the proliferation risk, a core designed to use MOX (Mixed Oxide) fuel is proposed, its performance investigated and the level of usability of the plutonium for non-peaceful applications assessed.

1. INTRODUCTION

Proliferation resistance has become one of the primary topics to be addressed if new nuclear energy systems are being developed as any nuclear system presents potential proliferation risks. Small Modular Reactor (SMR) systems could raise specific proliferation concerns mainly because they could be deployed in: i) remote areas, ii) small countries, iii) in large numbers, iv) in countries that are "newcomers" in nuclear industry and v) not only for electric generation (potable water production, process heat, etc.). In this sense, the whole SMR system requires specific attention in order to reduce the attractiveness of fissile material that could be used for nuclear weapons. In this work the level of proliferation attractiveness of MOX (Mixed Oxide) fuel loaded in a typical SMR core, based on pressurized light water technology (PWR), will be analyzed and discussed. It is obvious that the burning of plutonium in MOX fuels could be a useful non-proliferation measure to reduce the existing separated civil and military Pu worldwide stockpiles estimated in approximately 460 metric tons. [Polidoro, 2015].

2. ADDRRESSING FISSILE MATERIAL TYPE

The main nuclear material characteristics that need to be taken into account when considering a direct-use material in nuclear explosives are *bare spherical critical mass* (amount of material needed for achieving criticality), *heat generation* (amount of heat generated by the candidate weapons materials, potentially an issue for the stability of the chemical explosive and for the crystal structure of Pu), *spontaneous neutron emission rate* (amount of neutrons emitted by the nuclear material that can cause pre-initiation of the reaction-chain) and *radiation dose* (an issue for safe handling and storage of the device) [Cojazzi, 2012].

The categorization of fissile material considered here is based on the approached proposed by Pellaud [Pellaud, 2002] and focusing on spontaneous neutron emission. In the Pellaud's approach the usability of plutonium for building a nuclear weapon is based on the concentration of even-numbered Pu isotopes (mainly ^{240}Pu) and follows a coarse categorization in Super-grade (SG) , Weapon-grade (WG), Fuel-grade (FG), Reactor-grade (RG) and MOX-grade, as reported in Table 1. Pellaud argues that for verification purposes and to differentiate the proliferation resistance of Pu mixtures, it would be useful to define a category of Pu for which the technical difficulties to use it in a weapon are so high that it is "practically unusable". He gives a threshold of 30 wt% for ^{240}Pu. With that definition, plutonium contained in MOX fuel would be practically unusable for weapon purposes. However, it is questionable if ^{240}Pu content alone is a sufficient measure to quantify proliferation resistance. In fact, recently Bathke et. al., [Bathke, 2010] proposed a new approach for the categorization of fissile material that uses the so-called Figures of Merit in two variants (FOM1 and FOM2). As preliminary approach to the problem of proliferation resistance of SMR fuels and, in particular, the usability of plutonium for non-peaceful applications, the Pellaud categorization will be considered in the present study.

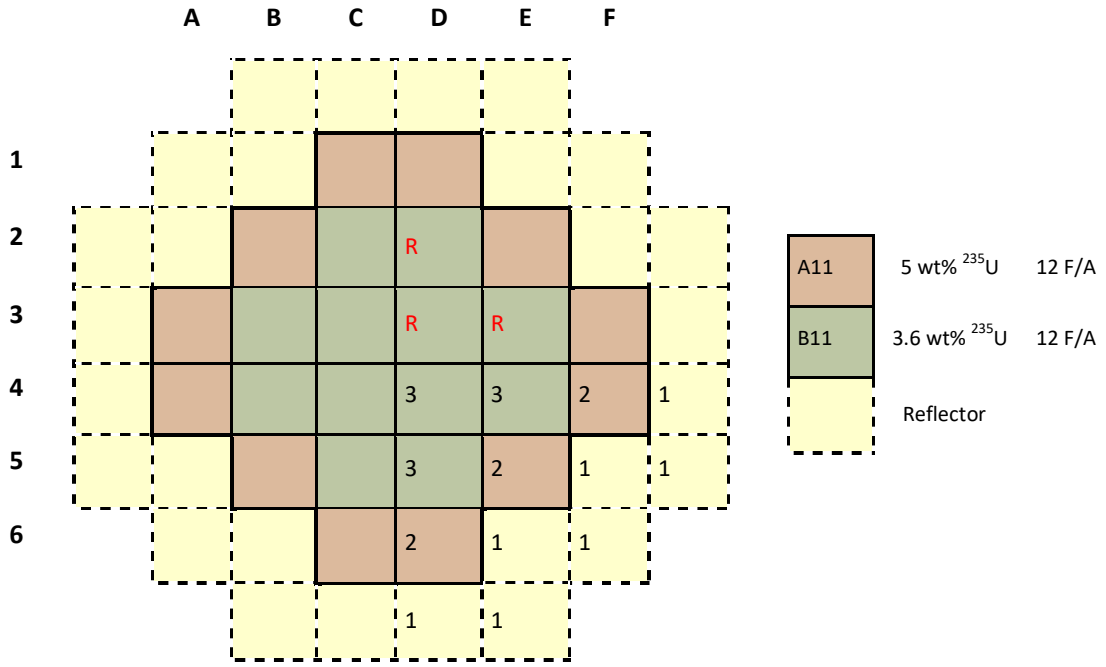
Table 1 – Pellaud's assessment of the usability of various Pu categories.

Category	^{240}Pu abundance range (wt%)	Usability for a nuclear weapon
Super-grade (SG)	< 3	Best quality
Weapon-grade (WG)	3 - 7	Standard material
Fuel-grade (FG)	7 - 18	Practically usable
Reactor-grade (RG)	18 - 30	Conceivably usable
MOX-grade	>30	Practically unusable

2. NEUTRONIC ANALYSIS OF A SMR CORE

2.1 Reference design

The reference core considered in the present study is that of a typical 150 MW thermal power pressurized water reactor designed for being operating with UO_2 fuel. The reactor consists of an array of 24 Fuel Assemblies (F/A), identical from the mechanical design point of view, loaded in the core accordingly to the scheme of Figure 1 (first core loading). These assemblies have mechanical and geometrical designs of a typical large power rated PWR while the active length is limited to 1.35m. The core density power is assumed to be 100 kW/l.



Legend: 1 = reflector, 2 & 3 = fuel assemblies, R = rodded fuel assemblies (12 assemblies).

Figure 1 – Core loading pattern at Beginning Of Life (BOL).

Fuel assemblies are of type 17 x 17, containing 264 fuel rods, as shown in Figure 2. Fuel rods are made of Zircaloy tubing containing UO_2 fuel pellets with enrichment in ^{235}U at 5 wt% while at Beginning-Of-Life (BOL) the inner part of the core is also loaded with 12 F/A at 3.6 wt% ^{235}U. The A_{11} fuel assembly contains around 173 kg of heavy metal with 94.95 wt% of ^{238}U and 0.05 wt% of ^{234}U (in the B_{11} F/A the percentages of these isotopes are 96.37 wt% and 0.03 wt%, respectively). Some assemblies contain fuel rods with gadolinium (Gd_2O_3) as burnable poison (integral burnable poison fuel pins); from 12 to 16 Gd-rods are included in the assembly with an absorber content of 8 wt%. A two-batch refueling scheme is adopted: the 12 F/As located in the inner part of the core are unloaded at the end of each burnup cycle (EOC) and replaced by the outer F/As with 5 wt% ^{235}U.

A total of 24 positions in the 17x17 array are equipped with guide thimbles, which are used as location for Rod Cluster Control Assemblies (RCCAs), while a central one is used for instrumentation purpose. The core has a shutdown system made up to 12 RCCAs (the 12 central F/As), each of them containing 24 absorber rods (Ag, In, Cd), over a length which covers nearly the complete active fuel length. The core is axially and

radially surrounded by a layer of reflector assemblies made up of structural materials (stainless steel, Zircaloy) and water; the total core height is of around 1.75 m including the axial reflectors (bottom and top) while the equivalent core diameter is of around 1.2 m.

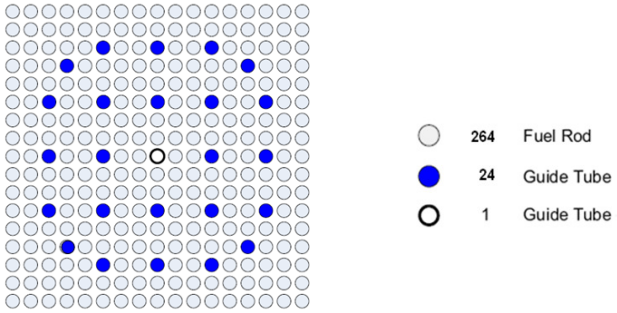


Figure 2 – Assembly fuel pattern.

The coolant contains boron as neutron absorber for controlling slow reactivity changes during operation at power (Xe-poisoning and burn-up effects) and for compensation of large reactivity changes during cool-down or heat-up phases. The main core and assembly data are reported in Table 2. Most of the data are deduced form available literature on SMRs [IAEA, 2007] or from the modeling of large PWR cores [RSE, 2011], [Global2011, 2011].

As in large PWR core, the use of burnable poison (G_2O_3) has the effect to reduce the reactivity excess at the beginning of life and thus the level of diluted boron in the coolant; as example, Figure 3 shows the impact of burnable poison on the value of k_∞ for the assembly type A_{11} along first burn-up cycle.

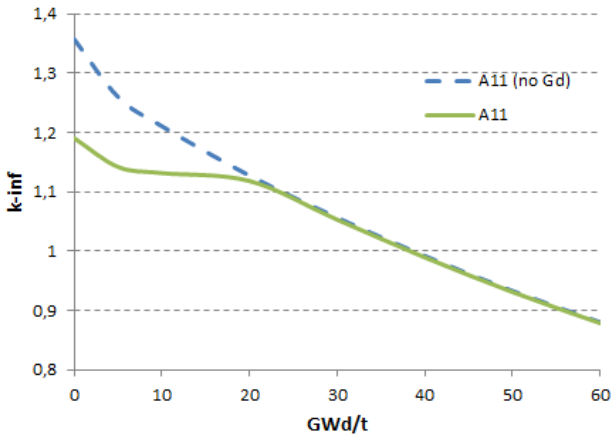


Figure 3 – Trend of reactivity along the first burn-up cycle for the fuel assembly A_{11}.

Table 2 – Core design data.

Parameter	Unit	Value	Ref.	Remarks
Total N° F/A. in the core	-	24	[IAEA,2007]	*See Fig. 1*
Total N° fuel rods	-	6336	[IAEA,2007]	
Fuel pattern	-	17 x 17	[IAEA,2007]	*See Fig. 2*
Fuel assembly pitch	cm	21.5	[RSE, 2011]	
Pin pitch	cm	1.26	[RSE, 2011]	
N° fuel pins per assembly	-	264	[IAEA,2007]	*See Fig. 2*
N° control rod pins per assembly	-	24	[RSE, 2011]	*See Fig. 2*
N° instrumentation tube per ass.	-	1	[RSE, 2011]	*See Fig. 2*
Active length	cm	135	[IAEA,2007]	
Ratio height/diameter	-	1.14	-	
Density power	kW/l	100	[IAEA,2007]	
Total thermal power	MWth	150	-	
Fuel pellet diameter	cm	0.819	[RSE, 2011]	
Cladding material	-	Zr-4	[IAEA,2007]	
Control rod & instr. data	-	-	[RSE, 2011]	
Fuel enrich. ^{235}U (@ BOL)	wt%	3.6 - 5	[RSE, 2011]	
Total fuel volume	m^3 (litres)	1.50 (1498)	-	
Core pressure	MPa	15.5	[RSE, 2011]	
Coolant flow rate	kg/s	721	-	*Scaled from large PWR cores*
Rated coolant mass	kg /(cm^2 h)	234	Calculated	
Inlet coolant temperature	°C	295	[RSE, 2011]	
Outlet coolant temperature	°C	330	[RSE, 2011]	
Average fuel temperature	°C	622	[RSE, 2011]	
Average moderator temp.	°C	313	[RSE, 2011]	
Burnable poison	-	Gd_2O_3	[RSE, 2011]	
N° of Gd_2O_3 pins per assembly	-	16 pins 8 wt% @ 5 wt% ^{235}U 12 pins 8 wt% @ 3.6wt% ^{235}U	-	*N. and wt% to limit reactivity excess @ BOL*
Reactivity control	-	RCCAs and diluted boron	[RSE, 2011]	
RCCAs (Ag, In, Cd)	wt%	80, 15, 5	[RSE, 2011]	

2.2 Steady state simulation of the UO_2 core

The analysis of the UO_2 core behavior in steady state conditions up to the equilibrium cycle have been carried out by using a state of art neutronic PWR core simulator. Main results are reported in the Figures 4 ÷ Figure 9 and Table 3; more specifically:

- Figure 4 shows the trend of critical boron (in ppm ^{10}B) in the core vs. burn-up (in GWd/t) at equilibrium cycle in hot full power conditions and with all RCCAs out (equilibrium Xenon);

- Figure 5 shows trends of axial relative power fraction and axial peak power distribution (in W/cm) at Beginning Of Equilibrium cycle (BOEC), at Middle Of Equilibrium Cycle (MOEC) i.e. 8 GWd/t, and at the End Of Equilibrium Cycle (EOEC) i.e. 14.9 GWd/t;

- Figure 6 ÷ Figure 8 show the assembly 2-D average relative power fraction and burn-up distribution (in GWd/t) at BOEC, MOEC and EOEC (results reported on ¼ core);

- Figure 9 shows the effective delayed neutron fraction (in pcm) vs. burn-up at the equilibrium cycle;

- Table 3 reports the values of reactivity coefficients (control rod worth, boron coefficient, Doppler, moderator temperature coefficient) in hot-full-power (HFP) and hot-zero-power (HZP) conditions at BOEC.

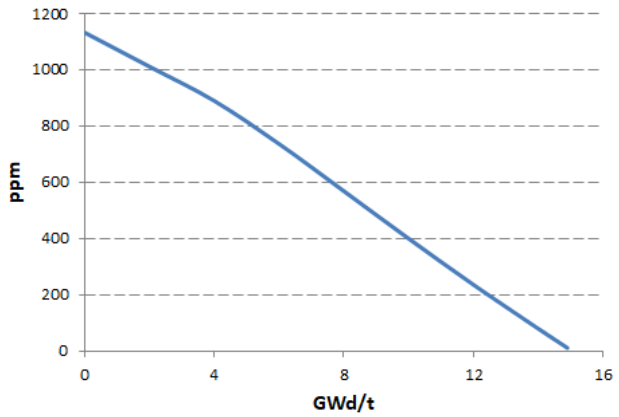


Figure 4 – Critical boron vs. burn-up at the equilibrium cycle

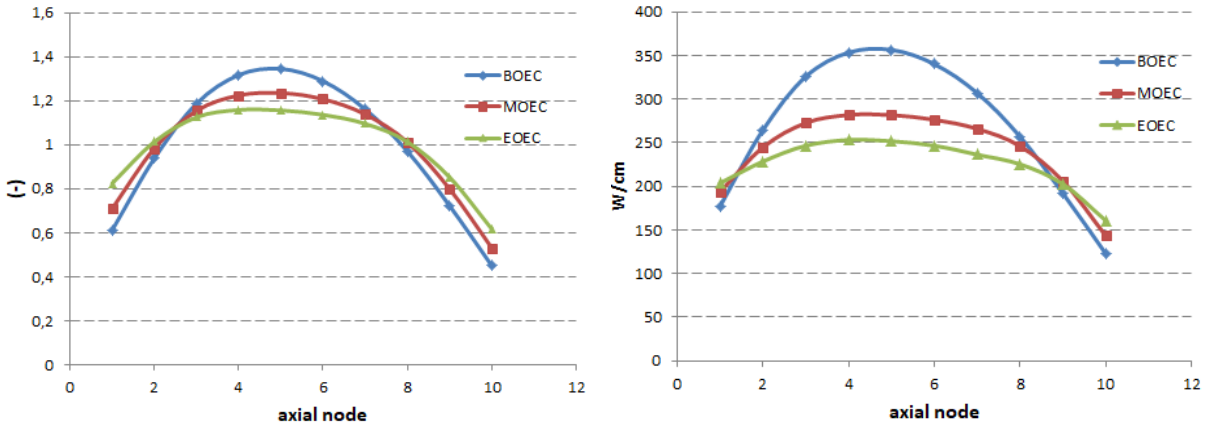


Figure 5 – Axial relative power fraction (left) and axial peak power distribution (right) at BOEC, MOEC and EOEC.

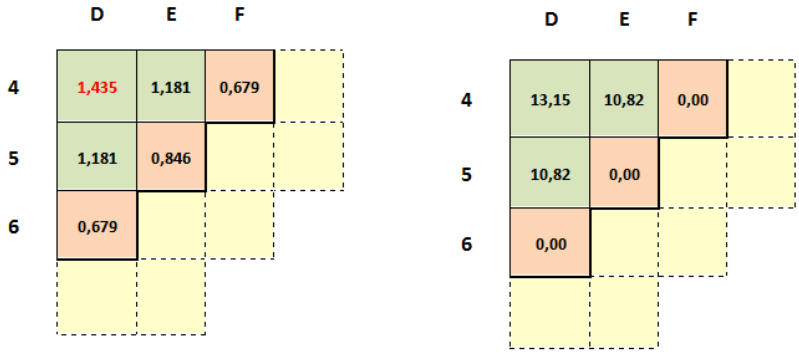


Figure 6 - Assembly 2-D average relative power fraction (left) and burn-up distribution (right) at BOEC.

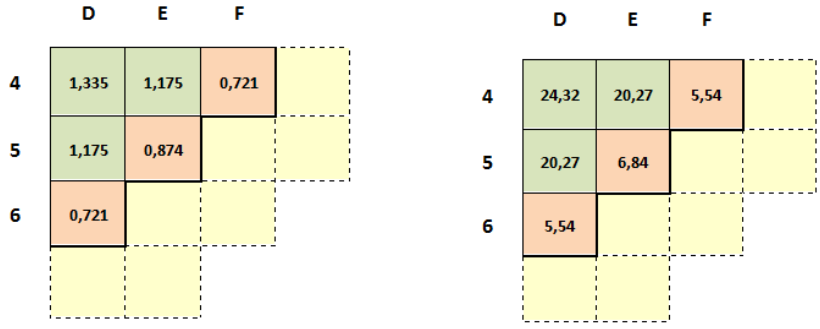


Figure 7 - Assembly 2-D average relative power fraction (left) and burn-up distribution (right) at MOEC.

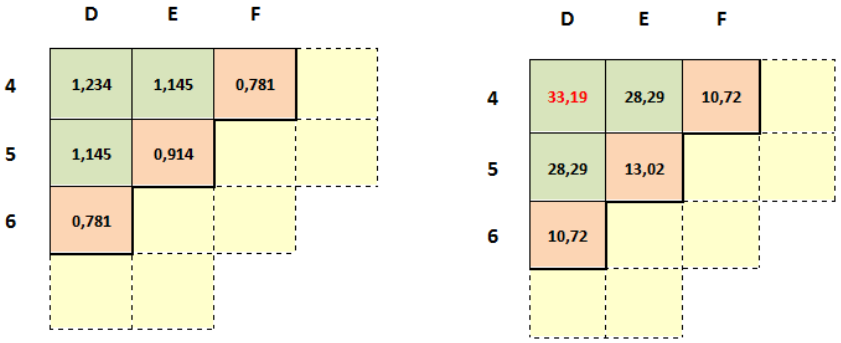


Figure 8 - Assembly 2-D average relative power fraction (left) and burn-up distribution (right) at EOEC.

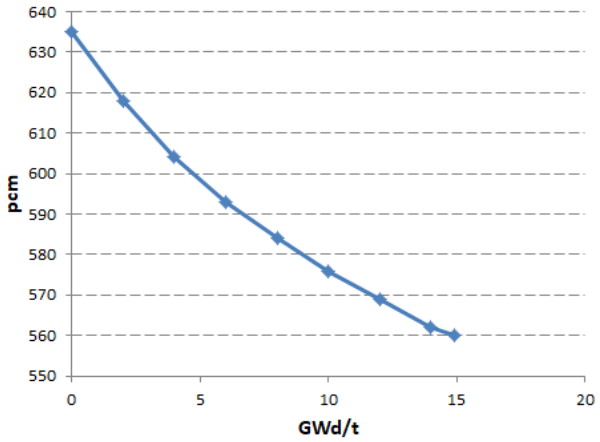


Figure 9 – Effective delayed neutron fraction vs. burn-up at the equilibrium cycle.

Table 2 – Reactivity coefficients evaluated at BOEC.

Reactor condition	Parameter	Unit	Value
HFP, ARI	Control rod worth (Δk/k)	(pcm)	-18717
HZP, ARI	Control rod worth (Δk/k)	(pcm)	-17824
HFP	Boron coefficient	(pcm/ppm ^{10}B)	-5.48
HFP	Doppler	(pcm/°F)	-1.51
HZP	MTC	(pcm/°F)	-10.50

Legend: ARI = all RCCAs inserted, MTC = Moderator temperature coefficient

The main outcomes from the numerical simulations of the UO_2-core are:

- the length of the equilibrium cycle is of 14.90 GWd/t corresponding to 468 Effective Full Power Days (EFPDs);

- the boron concentration required at BOEC to make the core critical is of 1132 ppm and linearly decreases with burn-up with a rate of around 75 ppm/GWd/t;

- the 2-D assembly normalized power distribution at different burn-up levels shows a reasonable trend, with a maximum peak power of 1.435 times the core average power reached at BOEC, in the assembly at location [D,4];

- the maximum linear power density remains under the value of 357 W/cm (value reached at BOL), i.e. below the thermal-hydraulic design data considered for typical PWR cores;

- the highest exposure level is reached in the central fuel assemblies at EOEC with 33.19 GWd/t, while the average core burn-up is of around 20.70 GWd/t;

- the effective delayed neutron fraction (β_{eff}) is equal to 635 pcm at BOEC and slightly decreases with burn-up;

- all the reactivity coefficients are negative with large margin of the RCCAs worth in HFP and HZP conditions.

2.3 Isotopic evolution and plutonium usability for the UO_2 core

The level of proliferation attractiveness of fuel in the core requires the analysis of the isotopic evolution of the fissile materials at each burn-up level. The isotopic evolution of uranium and plutonium isotopes at 1st cycle, 2nd cycle and at equilibrium cycle is reported in the following figures; more specifically:

- Figure 10 shows trend of mass (in kg) of ^{235}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu vs. burn-up for the 1st cycle.

- Figure 11 shows trend of mass (in kg) of ^{235}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu vs. burn-up for the 2nd cycle.

- Figure 12 shows trend of mass (in kg) of ^{235}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu vs. burn-up at equilibrium cycle.

As far as the evolution of the fissile isotopes it is possible to conclude that:

- at the equilibrium cycle around 54 kg of ^{235}U undergo fission in the core (30% of total).

- the amount of ^{239}Pu and ^{241}Pu increase with burnup in each cycle and at the equilibrium there is a net production in the core of around 17 kg (+14 kg for ^{239}Pu and +3 kg for ^{241}Pu).

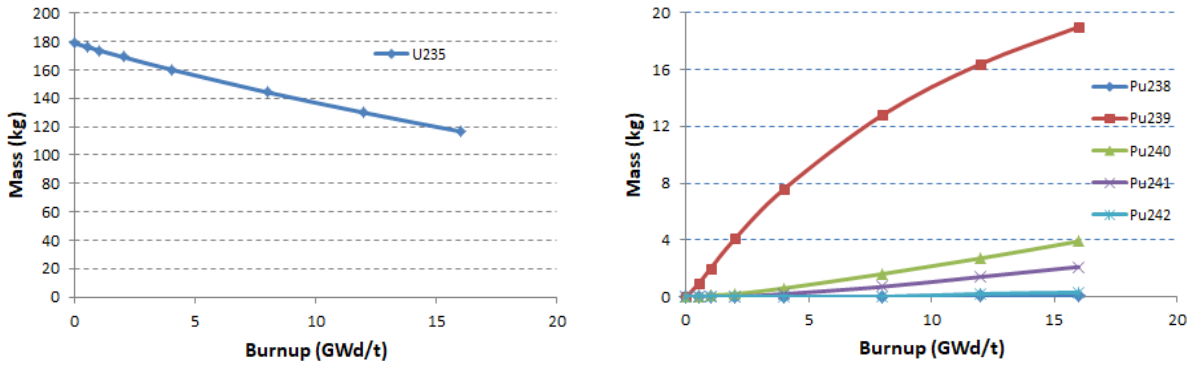


Figure 10 – Mass of ^{235}U (left) and of plutonium isotopes (right) vs. burn-up for the 1st cycle

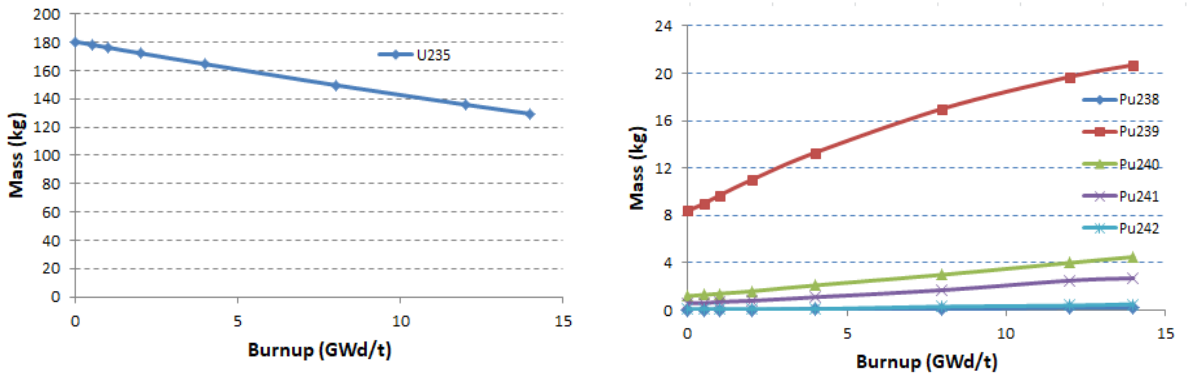


Figure 11 – Mass of ^{235}U (left) and of plutonium isotopes (right) vs. burn-up for the 2nd cycle.

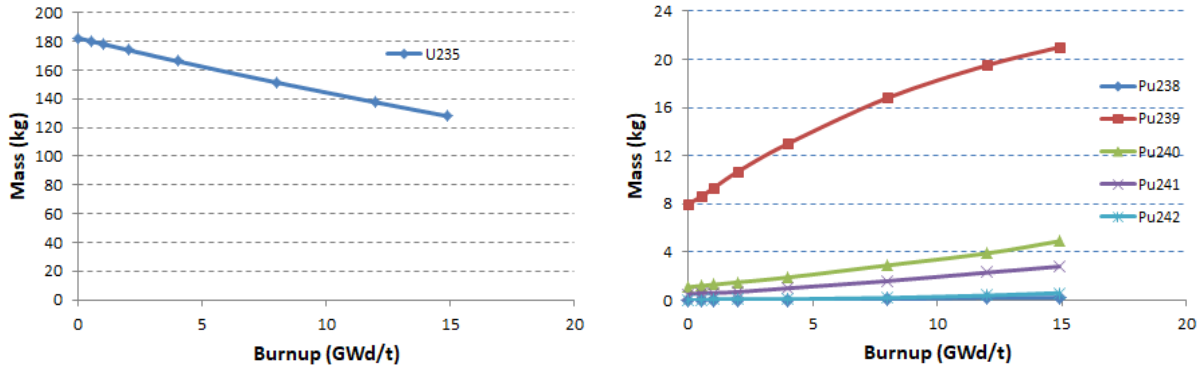


Figure 12 – Mass of ^{235}U (left) and of plutonium isotopes (right) vs. burn-up at equilibrium cycle.

Based on the categorization of fissile material proposed by Pellaud (^{240}Pu abundance), the plutonium usability during the 1st cycle, 2nd cycle and equilibrium cycle is summarized in Table 3 ÷ Table 5 and Figure 13 ÷ Figure 15.

Table 3 – Analysis of plutonium usability for the 1st cycle.

Burn-up (GWd/t)	Pu^{fiss}/Pu^{tot}	$^{240}Pu/Pu^{tot}$ (%)	Material Category
0	0	0	-
0.5	0.993	0.7	SG
1	0.974	2.6	SG
2	0.958	4.2	WG
4	0.932	6.8	WG
8	0.890	10.5	FG
12	0.857	13.2	FG
16	0.830	15.3	FG

Note. The amount of Pu in each category is: SG = 2.04 kg, WG = 5.73 kg, FG = 13.33 kg (total fissile Pu = 21.10 kg).

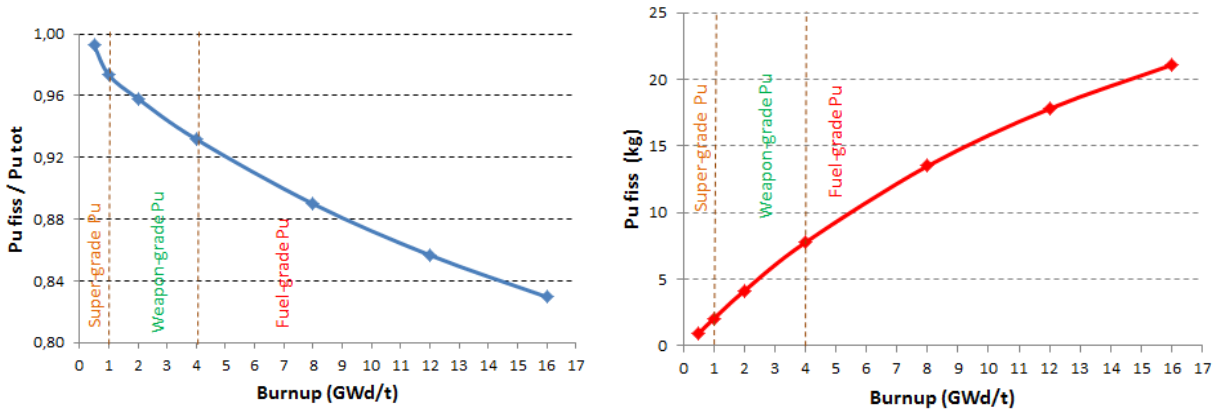


Figure 13 – Trend of ratio $Pu^{fissile}/Pu^{total}$ (left) and of total amount of fissile plutonium ($^{239}Pu+^{241}Pu$) (right) vs. burn-up for the 1st cycle.

Table 4 – Analysis of plutonium usability for the 2nd cycle.

Burn-up (GWd/t)	Pu^{fiss}/Pu^{tot}	$^{240}Pu/Pu^{tot}$ (%)	Material Category
0	0.874	11.8	FG
0.5	0.873	11.8	FG
1	0.873	11.8	FG
2	0.870	11.9	FG
4	0.865	12.3	FG
8	0.846	13.7	FG
12	0.827	15.0	FG
14	0.817	15.7	FG

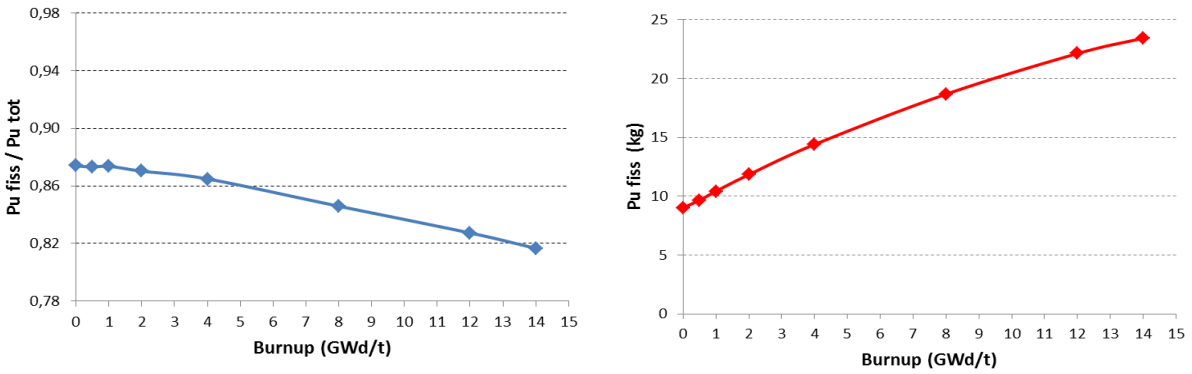


Figure 14 – Trend of ratio $Pu^{fissile}/Pu^{total}$ (left) and of total amount of fissile plutonium ($^{239}Pu+^{241}Pu$) (right) vs. burn-up for the 2nd cycle.

Table 5 – Analysis of plutonium usability at the equilibrium-of-cycle.

Burn-up (GWd/t)	Pu^{fiss}/Pu^{tot}	$^{240}Pu/Pu^{tot}$ (%)	Material Category
0	0.882	11.1	FG
0.5	0.881	11.2	FG
1	0.881	11.2	FG
2	0.879	11.3	FG
4	0.870	11.9	FG
8	0.851	13.4	FG
12	0.830	14.8	FG
14.9	0.815	15.8	FG

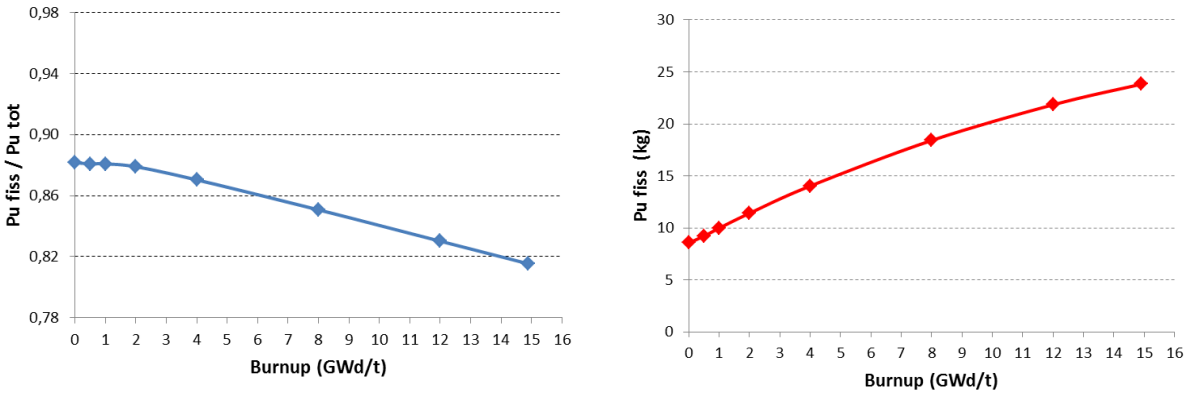


Figure 15 – Trend of ratio $Pu^{fissile}/Pu^{total}$ (left) and of total amount of fissile plutonium ($^{239}Pu+^{241}Pu$) (right) vs. burn-up at equilibrium of cycle.

In terms of proliferation risk, the UO_2 core is enable to produce nuclear material with high grade of usability (SG) for nuclear weapon within the first GWd/t of exposure. At 4 GWd/t core exposure in the first cycle, around 7.7 kg of fissile plutonium weapon-grade (WG)[1] are produce in the core, while the plutonium quality is downgraded to fuel-grade only above this exposure value. At the equilibrium cycle, only fuel-grade plutonium (FG) is produce in the core with a ratio $^{240}Pu/Pu^{tot}$ varying from 11.1% at BOEC to 15.8% at the EOEC. According to the Pellaud's fuel categorization, plutonium produced in the UO_2 core is practically usable for the realization of nuclear weapons.

[1] *6 kg plutonium weapon-grade is the amount of fissile material used for the realization of the first nuclear weapon of implosion type detonated at Nagasaki in August 1945.*

3 PROPOSAL FOR A PROLIFERATION RESISTANT SMR CORE

3.1 MOX core design

In order to face the proliferation risk, a modified design has been proposed for the SMR core by the use of MOX fuel. Starting from the initial core design, the original UO_2 fuel assemblies have been replaced by the MOX ones containing a mix of plutonium and ^{235}U as fissile materials, in different percentages, depending on their location in the core. In the MOX F/As the ^{240}Pu contents is set in such a way to assure that the fissile material is practically unusable for development of weapon devices, i.e. at each exposure level of the core the ratio $^{240}Pu/Pu^{tot}$ is always above 30% (MOX-grade fuel).

The weight fractions of plutonium isotopes included in the fuel at BOL are reported in Table 6; the isotope composition is representative of the plutonium produced in a traditional LWR fuel after an exposure of 45 GWd/t and 7 years of cooling/storage [CEA, 1997]. In Table 7, the weight fractions of fissile plutonium ($^{293}Pu+^{241}Pu$), total plutonium and ^{235}U loaded in the fuel at BOL are reported.

Provided a MOX assembly has only a modest plutonium concentration (less than about 10 wt%), a negative void coefficient is guaranteed in all normal operating core conditions. However, if the plutonium concentration is high enough the void coefficient will become positive and this will ultimately induce safety concerns. For that reason, in order to avoid any risk of positive reactivity void coefficient in the core, in case of generation of coolant voids, the maximum amount of plutonium loaded in the fuel has been limited to 14 wt% [2]. The maximum amount of plutonium T to be loaded in the fuel has been evaluated on the base of the following correlation that approximates the limit content for any vector, as a function of mass concentration C_i of 4 isotopes ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu [CEA, 1997]:

$$T\ (wt\%) = 0.1156845\ C_{239\ Pu} + 0.214839\ C_{240\ Pu} + 0.0222565\ C_{241\ Pu} + 0.220139\ C_{242\ Pu}$$

[2] *The void reactivity coefficient measures how much reactivity changes when the moderator density decreases when bubbles of steam form in a LWR. It is usual for the nuclear designer to ensure that the void coefficient is always negative, so that any voidage decreases reactivity and provides negative feedback. The voiding is a complex phenomenon whose effects are related to a spectrum variation disturbing drastically the competitions between productions and absorptions of some isotopes having strong resonances. The variation of contribution of each isotope is as follows: the most important contributions come from ^{238}U (always negative and a function of content), from ^{240}Pu (always positive and not very depending on the content), from ^{239}Pu (negative, or close to zero for high concentrations), from ^{242}Pu (positive or equal to zero, very depending on the isotopic concentration). The highly negative contribution of ^{238}U is due to an important increase of absorption in the 10 MeV – 1 KeV energy range, which compensates amply the vanishing of the resonant capture (5 – 200 eV) when the spectrum becomes harder, due to voiding. The ^{240}Pu has a capture centered on the thermal domain with a large resonance at 1 eV. The spectrum hardening reduces to zero this capture while increasing the production over 1 MeV (decrease of $\alpha = \sigma_c / \sigma_f$ and increase of v number of neutrons emitted per fission), thus giving a highly positive contribution. Sensitivity studies have been performed in order to determine a limit for the total plutonium content in the MOX fuel, as regard to the total voidage of the core. This conservative limit varies from 12.5 wt% (vector with very high content of fissile plutonium: 90%) to 15 wt% (vector with very degraded plutonium: 48% fissile plutonium), i.e. the more the plutonium is degraded, the higher is the limit content. However, the mentioned problems occur only at high voiding conditions, i.e. at beyond 60% of voids.*

At BOL the core is loaded with 12 F/As of the A_{11} type, each one containing around 171.3 kg of heavy metal; the weight fractions of the isotopes ^{234}U, ^{235}U, ^{238}U, ^{238}Pu ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu are 0.01, 1.03, 84.82, 0.42, 6.79, 4.24, 1.41 and 1.2, respectively. For the 1st cycle only, 12 F/As of the B_{11} type are also loaded; each fuel assembly contains 171.3 kg of heavy metal while the weight fractions of the isotopes ^{234}U, ^{235}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu are 0.01, 1.09, 89.71, 0.28, 4.41, 2.76, 0.92 and 0.78. A two-batch refueling scheme is still adopted for the MOX core.

The MOX assemblies loaded in the core have same mechanical and geometrical designs of those considered in the original design and the only modifications concerns the introduction of pins containing burnable poison (Gd_2O_3) for the reduction of reactivity excess at BOL (16 pins @ 2 wt% Gd_2O_3 for A_{11} assemblies and 12 pins @ 2 wt% Gd_2O_3 for B_{11} assemblies). Thermal-hydraulic operating conditions, rated power, reactivity control systems and refueling scheme are the same of the UO_2 reference core.

Table 6 – Weight fractions of plutonium isotopes in the fuel.

^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
3.0	48	30	10	8.5	0.5

Table 7 – Weight fractions of plutonium and uranium in the fuel at BOL (1st cycle).

Cycle #	F/A type	Pu^{fiss} (wt%)	Pu^{tot} (wt%)	^{235}U (wt%)
1	A11	8.2	14.1	1.03
	B11	5.3	9.1	1.09

It should be pointed out that the use of identical mechanical design avoids issues of thermal-hydraulic and mechanical handling incompatibility. However, as it is well known, the presence of MOX fuel affects the design characteristics of the core and its performances. In fact, MOX fuel differs from the UO_2 fuel for the fact that the fissile materials are mainly ^{239}Pu and ^{241}Pu, rather than ^{235}U. Plutonium and uranium have fundamentally different nuclear cross sections and this determines different performances of the reactor core. Plutonium has a higher thermal absorption cross section $\sigma_{th,ab}$ and fission cross section $\sigma_{th,f}$, more neutrons per fission ν , a larger energy per fission E_f, and a smaller delayed neutron fraction β. Plutonium isotopes ^{239}Pu, ^{240}Pu and ^{242}Pu show strong resonance peaks in the near thermal region. As a result, the neutron spectrum in MOX fuel is hardened, i.e. at the same power level, the thermal neutron flux is much lower than that in the uranium fuel. All these different nuclear properties have an impact on the neutron spectrum, reactivity coefficients and absorber effectiveness. In particular, the smaller delayed neutron fraction results in changes in the kinetic response of the reactor, with the reactor responding more rapidly to reactivity changes.

3.2 Steady state core simulation of the MOX core

The analysis of the MOX core behavior in steady state conditions up to the equilibrium cycle have been carried out by using a state of art neutronic PWR core simulator. Main results are reported in the Figure 16 ÷ Figure 22 and Table 8; more specifically:

- Figure 16 shows the trend of critical boron (in ppm ^{10}B) in the core vs. burn-up (in GWd/t) at equilibrium cycle, in hot full power conditions and with all RCCAs out (equilibrium Xenon) (trend of critical boron for the UO_2 core is also reported for comparison purpose);

- Figure 17 shows trends of axial relative power fraction and axial peak power distribution (in W/cm) at BOEC, at MOEC i.e. 8 GWd/t and at EOEC i.e. 13.08 GWd/t;

- Figure 18 reports the comparison of the axial peak power distributions at BOEC for the UO_2 core and MOX core;

- Figure 19 ÷ Figure 21 show the assembly 2-D average relative power fraction and burn-up distribution (in GWd/t) at BOEC, MOEC and EOEC (results reported on ¼ core).

- Figure 22 shows the effective delayed neutron fraction (in pcm) vs. burn-up at equilibrium cycle;

- Table 8 reports the values of reactivity coefficients (control rod worth, boron coefficient, Doppler, moderator temperature coefficient) evaluated in hot-full-power (HFP) and hot-zero-power (HZP) conditions at BOEC (in the same table reactivity coefficients for the UO_2 core are also reported for comparison purpose).

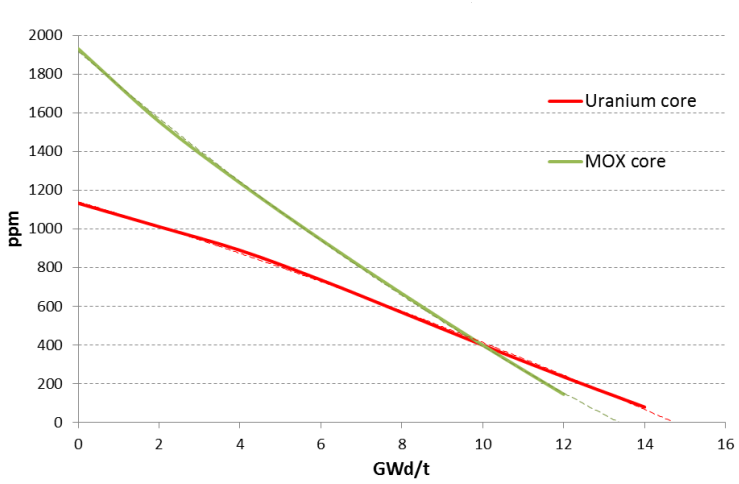


Figure 19 – Trends of critical boron vs. burn-up at equilibrium cycle for the MOX core and comparison with the trend for the UO_2 core.

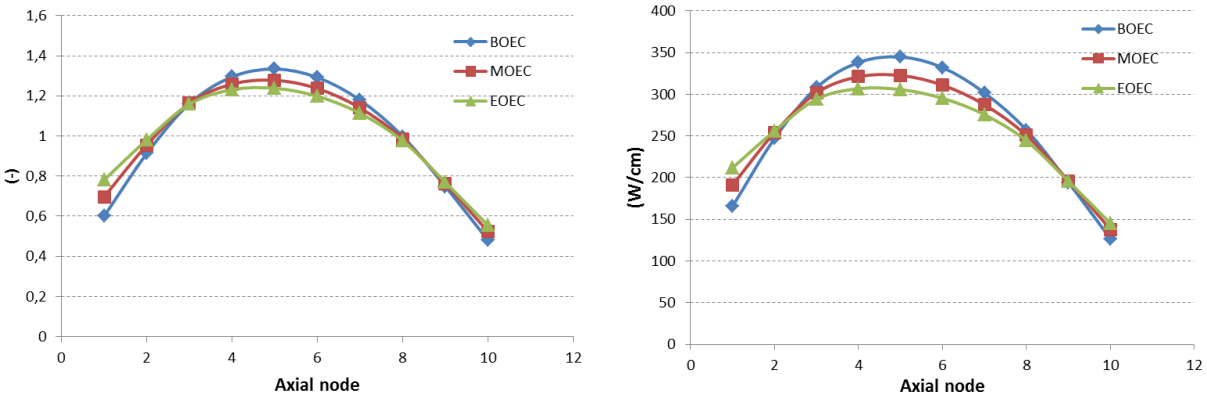


Figure 20 – MOX core: axial relative power fraction (left) and axial peak power distribution (right) at BOEC, MOEC and EOEC.

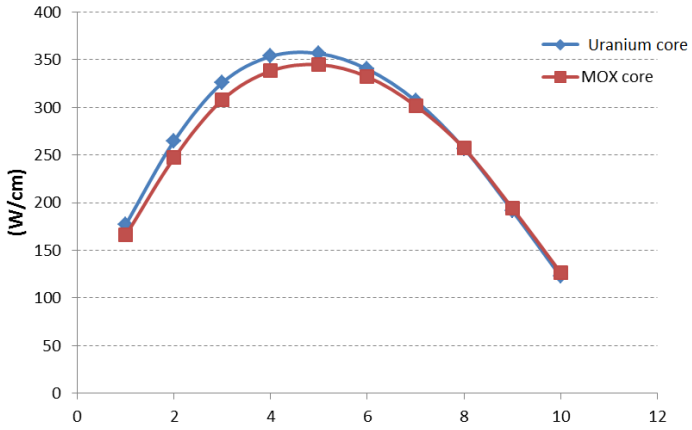


Figure 21 – Axial peak power distributions at BOEC for the MOX core and comparison with the trend for the UO_2 core.

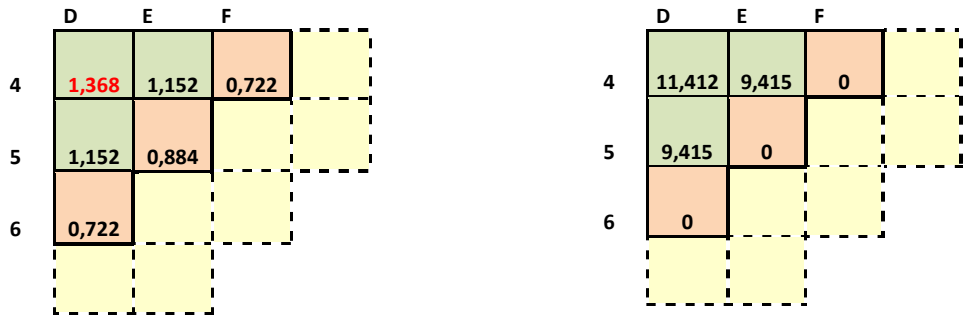


Figure 22- MOX core: assembly 2-D average relative power fraction (left) and burn-up distribution (right) at BOEC.

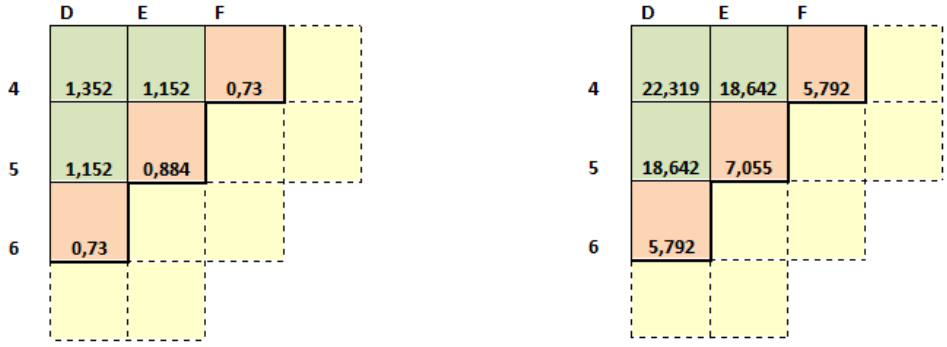


Figure 23- MOX core: assembly 2-D average relative power fraction (left) and burn-up distribution (right) at MOEC.

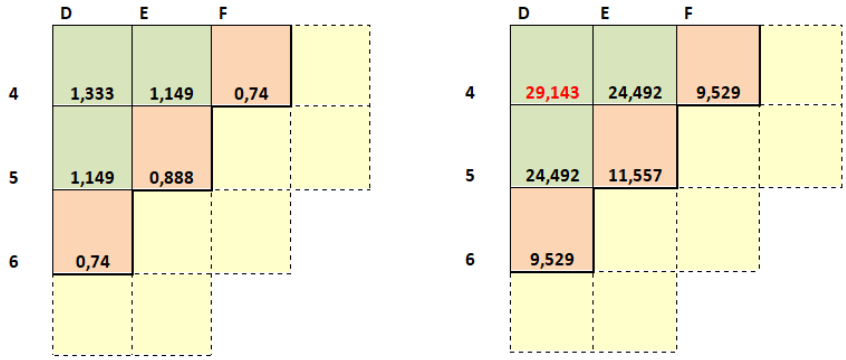


Figure 24- MOX core: assembly 2-D average relative power fraction (left) and burn-up distribution (right) at EOEC.

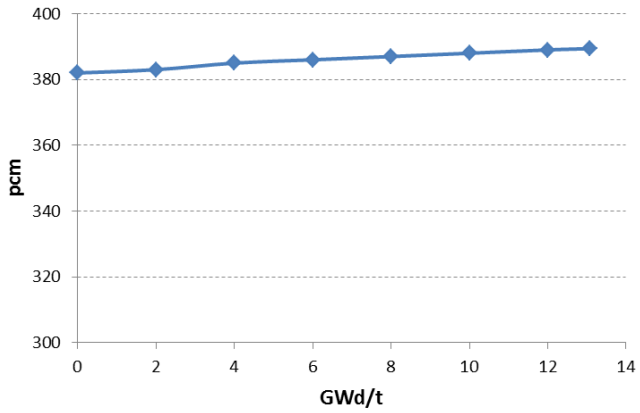


Figure 25 – MOX core: effective delayed neutron fraction vs. burn-up at the equilibrium cycle.

Table 8 – Comparison of reactivity coefficients for the MOX core and UO_2 core at BOEC.

Reactor condition	Parameter	Core design		$\Delta_{MOX-UO2}$
		MOX	**UO_2**	
HFP, ARI	Control rod worth (Δk/k) (pcm)	17987	18717	-730
HZP, ARI	Control rod worth (Δk/k) (pcm)	16989	17824	-835
HFP	Boron coefficient (pcm/ppm ^{10}B)	-1.79	-5.48	+3.69
HFP	Doppler (pcm/°F)	-1.59	-1.51	-0.08
HZP	MTC (pcm/°F)	-10.70	-10.50	-0.2

Legend: ARI = all RCCAs inserted, MTC = Moderator temperature coefficient

The main outcomes from the numerical simulations of the MOX-core are:

- the equilibrium cycle length is of 13.08 GWd/t corresponding to 358 EFPDs, i.e. the MOX core shows a shorter equilibrium cycle length in comparison with the UO_2 core (-110 EFPDs);

- the boron concentration required at BOEC to make the core critical is of 1931 ppm and linearly decreases with burn-up; in comparison with the UO_2 core, a higher contents of ^{10}B in the coolant is needed for the MOX core (+ 799ppm at BOEC);

- the 2-D assembly normalized power distribution at different burn-up levels shows a reasonable trend with a maximum peak power of 1.368 times the core average power reached at BOEC in the assembly at location [D,4] (1.435 in the UO_2 core);

- the maximum linear power density remains under the value of 345 W/cm @ BOEC (357 W/cm in the UO_2 core);

- the highest exposure level is reached in the central fuel assemblies at EOEC with 29.14 GWd/t, while the average core burn-up is of 18.2 GWd/t (33.19 GWd/t and 20.7 GWd/t for the UO_2 core, respectively);

- the use of MOX fuel in the core has an impact on the value of β_{eff} being equal to 385 pcm at BOEC (635 pcm for UO_2 core);

- all the reactivity coefficients are negative; however the use of MOX fuel has the effect to reduce the value of boron coefficient (from -5.48 pcm/ppm ^{10}B to – 1.79 pcm/ppm ^{10}B) as well as the control rod worth at HFP and HZP conditions (-730 pcm and -835 pcm, respectively). Doppler and MTC coefficients are not affected by the use of MOX in the core.

3.3 Isotopic evolution and plutonium usability for the MOX-core

The isotopic evolution of ^{235}U and plutonium for the 1st cycle and at equilibrium cycle is reported in the following figures:

- Figure 26 shows trend of mass (in kg) of ^{235}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu vs. burn-up for the first cycle.
- Figure 27 shows trend of mass (in kg) of ^{235}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu vs. burn-up at equilibrium cycle.

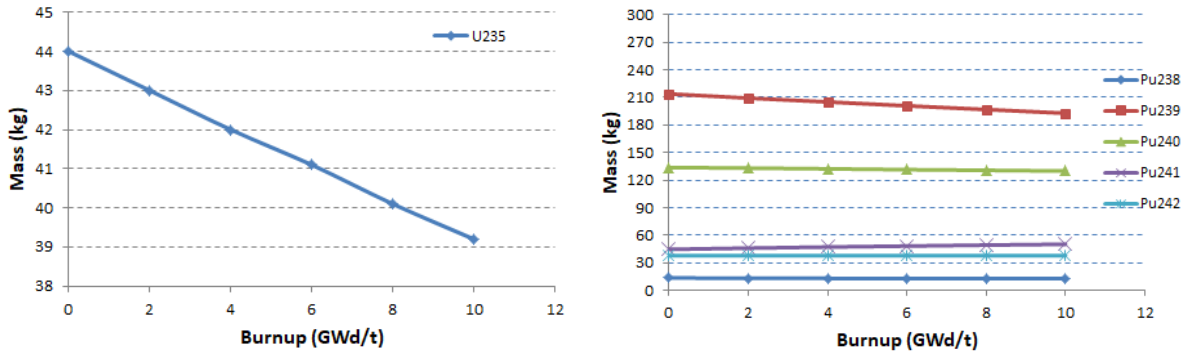


Figure 26 – MOX core: mass of ^{235}U (left) and of plutonium isotopes (right) vs. burn-up for the 1st cycle.

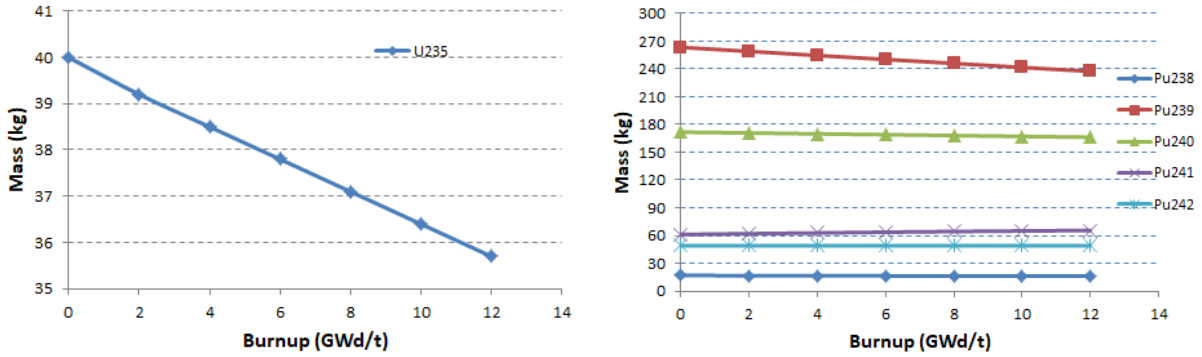


Figure 27 – MOX core: mass of ^{235}U (left) and of plutonium isotopes (right) vs. burn-up at equilibrium cycle.

From the analysis of the figures results:

- around 5 kg of ^{235}U undergo fission in the core (11% of total);

- the amount of fissile isotope ^{239}Pu decreases with burnup (-26 kg at equilibrium cycle) while the other fissile isotope ^{241}Pu slightly increases with burnup (+ 4.4 kg at equilibrium cycle); at the EOEC a net reduction of total fissile plutonium mass is obtained in the MOX core (-21.6 kg).

Based on the categorization of fissile material proposed by Pellaud (^{240}Pu abundance) the plutonium usability during the 1st and at equilibrium cycle is summarized in the following Table 9 ÷ Table 10 and Figure 28 ÷ Figure 29. As far as the proliferation risk, the MOX core presents, at each exposure level of the 1st cycle as well as at the equilibrium cycle a ^{240}Pu abundance higher than 30%, i.e. the plutonium is always of MOX-Grade type; in addition, this abundance is stable with the exposure, as shown in the Figure 30 (in the same figure the ratio Pu^{240}/Pu^{tot} vs. burn-up for the UO_2 cores is also reported for comparison purpose). This condition would assure that the plutonium contained in the MOX core is practically unable for the development of nuclear weapons.

Table 9 – Analysis of plutonium usability for the 1st cycle of the MOX core

Burn-up (GWd/t)	Pu^{fiss}/Pu^{tot}	$^{240}Pu/Pu^{tot}$ (%)	Material Category
0	0.583	30.1	MOX-Grade
2	0.581	30.3	MOX-Grade
4	0.579	30.4	MOX-Grade
6	0.578	30.4	MOX-Grade
8	0.576	30.6	MOX-Grade
10	0.574	30.6	MOX-Grade

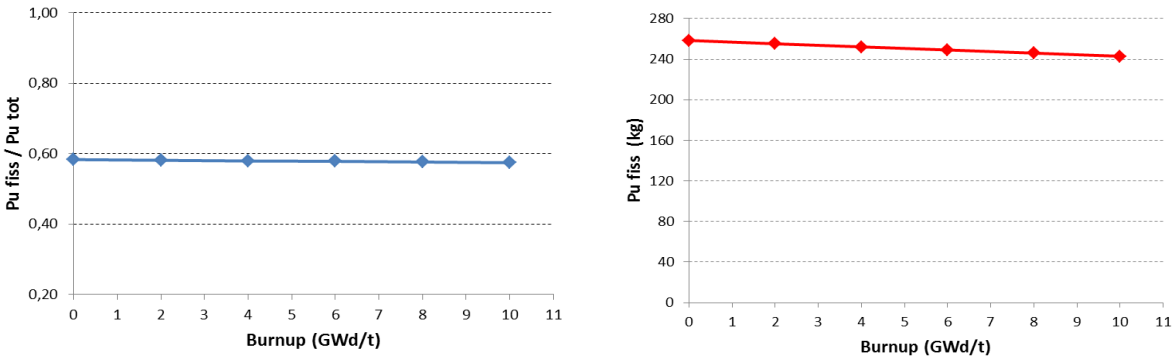


Figure 28 – MOX core: trend of the ratio Pu^{fiss}/Pu^{tot} (left) and of total amount of fissile plutonium ($^{239}Pu+^{241}Pu$) (right) vs. burn-up for 1st cycle.

Table 10 – Analysis of plutonium usability at equilibrium cycle for MOX core.

Burn-up (GWd/t)	Pu^{fiss}/Pu^{tot}	$^{240}Pu/Pu^{tot}$ (%)	Material Category
0	0.577	30.5	MOX-Grade
2	0.576	30.6	MOX-Grade
4	0.574	30.7	MOX-Grade
6	0.572	30.8	MOX-Grade
8	0.571	30.9	MOX-Grade
10	0.569	31	MOX-Grade
12	0.567	31.1	MOX-Grade

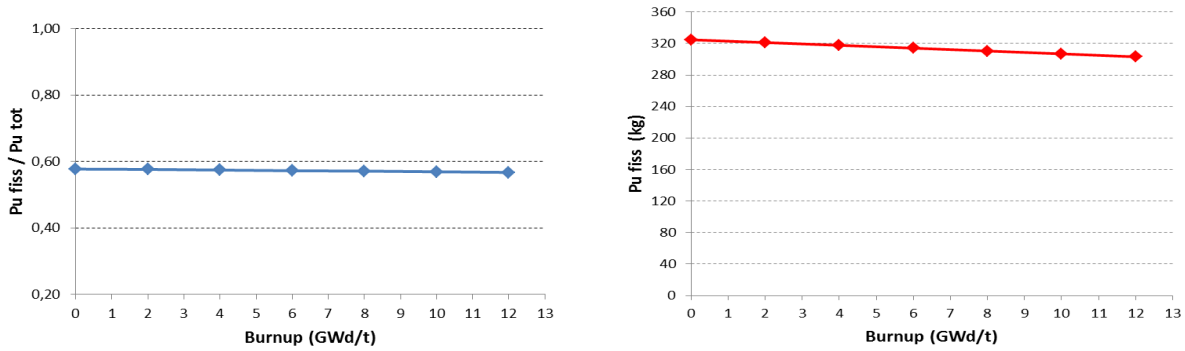


Figure 29 – MOX core: trend of the ratio Pu^{fiss}/Pu^{tot} (left) and of total amount of fissile plutonium ($^{239}Pu+^{241}Pu$) (right) vs. burn-up at the equilibrium cycle.

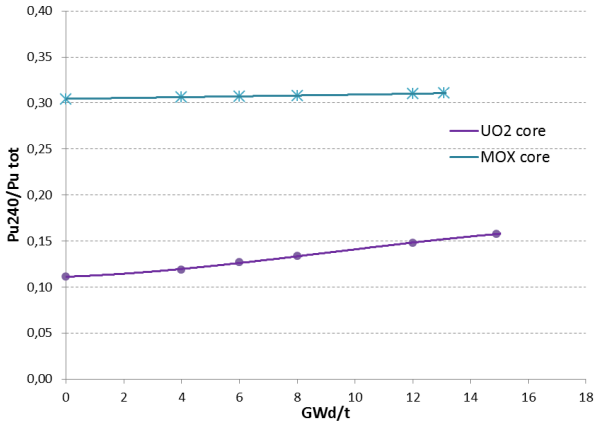


Figure 30 – Comparison of the ratio $^{240}Pu/Pu^{tot}$ vs. burnup for UO_2 core and MOX core (equilibrium cycle).

3.4 Sensitivity analysis

As mentioned in the section 3.1, in order to avoid any risk of positive void coefficient in the core, the maximum amount of plutonium in the fuel have to be limited. Conservative limits vary from 12.5 wt% to 15 wt% depending on the level concentration of fissile plutonium. In order to explore the feasibility of a core with lower plutonium content, a parametric study was performed by considering the Pu concentration to 12, 10, 8, 4 wt% while preserving, as much as possible, the cycle length at equilibrium. The isotopic compositions of the MOX core considered in section 3.1 (reference MOX case) and those of the additional cases considered in the sensitivity analysis are reported in the following Table 11 (the new cases are named #1, #2, #3 and #4, respectively). In the same Table 11, the assessment of plutonium usability at the EOEC for each case is also reported.

Table 11 – Assessment of plutonium usability of MOX cores with different Pu content.

Case	BOEC			$^{240}Pu/Pu^{tot}$ (EOEC) (%)	Material category
	Pu^{tot} (wt%)	Pu^{fiss} (wt%)	^{235}U (wt%)		
Ref. MOX	14	8.2	1.03	31	MOX-Grade
#1	12	7.0	2.0	31	MOX-Grade
#2	10	5.8	3.0	31	MOX-Grade
#3	8	4.7	3.6	31	MOX-Grade
#4	4	2.3	4.6	29	Reactor-Grade

As it can be deduced from Table 11, up to a Pu content of around 8 wt%, the fissile material in the core can be still categorized as reactor-grade; it is clear that the possibility to use a Pu concentration lower than 14 wt% would also reduce the impact of MOX fuel on the safety parameters of the core.

4. CONCLUSIONS

The aim of the present study was to identify the level of proliferation attractiveness of fuel loaded in a SMR core based on pressurized light water technology (PWR). A core designed to use MOX (Mixed Oxide) fuel was proposed, its performance investigated and the level of usability of the plutonium for non-peaceful applications assessed. The results of the numerical analyses show as at the equilibrium cycle the plutonium contained in the core is always of MOX-Grade type, i.e. the plutonium would be practically unusable for the realization of nuclear weapons. In order to avoid any risk of positive reactivity void coefficient in the core, the maximum amount of plutonium loaded in the fuel was limited for safety reasons to 14 wt%; however, the categorization of the fissile material as MOX-Grade can still be guaranteed if lower weight fractions of plutonium in the core are taken into account.

The results here presented are to be considered as preliminary and an optimization process on the core design should be envisaged in order to achieve, for instance (list not exhaustive):

- longer equilibrium cycle length (maximize fuel utilization);
- lower boron level in the core for reactivity control during normal operation (reducing safety concerns related to the boron dilution accidental);
- more flattened axial power distribution along the cycle;
- lower axial peak power value (increasing safety parameters);
- better neutronic economics;

This optimization could be pursued by modification of the core design in terms of i) axial differentiation of the burnable poison content in the fuel rod, ii) axial differentiation of ^{235}U enrichment in the fuel rods, iii) use of RCCAs system during the reactor operation to compensate the low boron contents in the coolant, iv) design of specific axial and radial reflectors to reduce the neutron leakages from the core, v) adequate fuel reloading scheme, etc.

NOMENCLATURE

ARI	All RCCAs In
ARO	All RCCAs Out
BOC	Beginning-Of-Cycle
BOEC	Beginning-Of-Equilibrium-Cycle
BOL	Beginning-Of-Life
MOC	Middle-Of-Cycle
MOEC	Middle-Of-Equilibrium-Cycle
EOC	End-Of-Cycle
EOEC	End-Of-Equilibrium-Cycle
EFPDs	Effective Full Power Days
F/A	Fuel Assembly
HFP	Hot Full Power
HZP	Hot Zero Power
MOX	Uranium-plutonium mixed oxide fuel
MTC	Moderator Temperature Coefficient
PWR	Pressurized Water Reactor
RCCA	Rod Cluster Control Assembly
SMR	Small Modular Reactor
SG-Pu	Super Grade Plutonium
WG-Pu	Weapon Grade Plutonium
FG-Pu	Fuel Grade Plutonium
RG-Pu	Reactor Grade Plutonium

REFERENCES

[Polidoro, 2015] F.Polidoro and F.Parozzi. *Settant'anni con la bomba*. Le Scienze, N. 563, July 2015.

[Cojazzi, 2012] M. Cojazzi et. al., *Material type and safeguardability considerations for innovative sodium fast reactors fuel including different minor actinides composition,* INMM-53rd Annual meeting, Orlando, Florida, USA, 2012.

[Pellaud, 2002] B. Pellaud, *Proliferation Aspects of Plutonium Recycling,* J. Nuclear Material Management, Vol. XXXI, No. 1, Fall, 2002.

[Bathke, 2010] C. G. Bathk et al., *The attractiveness of Materials in Advanced Nuclear Fuel Cycles for Various Proliferation and Theft Scenarios,* LA-UR-10-07282, 2010.

[IAEA, 2007] *Status of Small Reactor Designs Without On-Site Refuelling*, IAEA Report TEC-DOC-1536, Vienna, 2007

[RSE, 2010] F. Polidoro, G. Vimercati and E. Corsetti, *Modellazione del nocciolo di un reattore PWR,* RSE report No. 11000412, 2011.

[Global2011, 2011] F. Polidoro, G. Vimercati and E. Corsetti, *Preliminary Analysis of a Large 1600MWe PWR Core Loaded with 30% MOX Fuel,* Proceedings of GLOBAL2011, Makuhari, Dec. 11-16, Japan, 2011.

[CEA, 1997] S. Aniel et. al., *Evaluation of the maximum content of a MOX-fueled pressurized water reactor versus isotopic composition with respect to the void coefficient,* IAEA report TEC-DOC-941, Vienna, 1997