FEASIBILITY OF FUEL CYCLE BASED ON TH-MOX FUEL IN SMALL MODULAR REACTORS

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ABSTRACT

Uranium resources are secure for a long time, but prices are likely to be substantially higher after 2020. Nowadays, conventional industry, built on uranium, is watching with interest to thorium that could represent an alternative to uranium as fuel. Moreover, Thorium-MOX fuels, designed to be used in conventional LWRs, can utilize plutonium from spent fuel and weapons dismantling in order to provide a new option for reducing civil and military plutonium stocks. In this context, the sustainability of a fuel cycle based on thorium-plutonium mixed oxides in a Small Modular Reactor, is analyzed by using state-of-art neutronic numerical models. The work, in particular, analyzes the behavior of a SMR core fueled with Th-MOX fuel assemblies, and compares it with the analogous behavior of a SMR core fueled with conventional MOX. Main advantages and drawbacks of this option are also highlighted.

INTRODUCTION

Large stockpiles of civil plutonium have been accumulated in the world in different countries as consequence of nuclear power programs. There is a serious public and political concern in the world about the misuse of plutonium for nonconventional application and also for the accidental release of highly radiotoxic material into the environmental. One alternative for the management of plutonium is to incinerate it in power reactors. But if the plutonium is fueled in reactors in the form of uranium-plutonium mixed oxide (MOX), new plutonium is generated as consequence of the fertilization of ²³⁸U isotope. A possible solution to this problem is to incinerate plutonium in combination with thorium in the so called Th-MOX fuels. In fact, the thorium cycle produces ²³³U which from a non-proliferation point of view, is preferable to plutonium for two reasons. Firstly, it is contaminated with ²³²U, which decay to give highly active daughter products that would make handling and diversion difficult; secondly, the ²³³U could be denatured by adding some 238 U to the thorium, in such away to be sufficient to denature the 233 U, but not so much to produce a significant amount of fissile plutonium [1].

Several recent studies have analyzed the possibilities of Pu recycling in large PWR cores fueled with Th-MOX [2, 3]. 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

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enhance the capability to utilize plutonium from spent fuel and weapons dismantling, so providing a new option for reducing civil and military plutonium stocks [4]. In this context, the sustainability of a fuel cycle based on thorium-plutonium mixed oxides in a Small Modular Reactor, is analyzed by using stateof-art neutronic numerical models. The work, in particular, analyzes the behavior of a SMR core fueled with Th-MOX fuel assemblies, and compares it with the analogous behavior of a SMR core fueled with conventional MOX [5,6]. Main advantages and drawbacks of this option are highlighted.

BENEFITS AND CHALLENGES DERIVING FROM THE USE OF TH-MOX FUELS

In a scenario with progressive reduction of uranium natural resources, the use of thorium in the form of mixed oxide with plutonium as nuclear fuel, can results in several benefits as below described.

First of all, thorium is 3 to 4 times more abundant than uranium and it is widely distributed in nature as an easily exploitable resource in many countries. Unlike natural uranium, which contains ~0.7 wt% of fissile isotope ²³⁵U, natural thorium does not contain any fissile material and is made up of the fertile isotope ²³²Th only. Hence, thorium-based fuels (oxide or carbide), can be utilized in combination with other fissile material as ²³⁹Pu in nuclear reactors for power generation as well as for conversion to fissile isotope ²³³U, thereby enlarging the fissile material resources. Thorium fuels, therefore, complement uranium fuels and ensure long term sustainability of nuclear power¹.

Secondly, thorium fuel has a more favorable in-core behavior compared to MOX, due to its better material properties including a higher thermal conductivity, a significant higher melting point, a lower expansion coefficient, and a lower fission-gas release [7].

Thirdly, Th-based fuels and related fuel cycles have intrinsic proliferation-resistance due to the formation of ^{232}U isotope via (n, 2n) reactions with ^{232}Th , ^{233}Pa and ^{233}U . The half-life of ^{232}U (which emits α -particles) is of around 74 years and the daughter products have very short half-life and some of

¹ Thorium is a better fertile material than ^{238}U in thermal reactors thanks to its higher absorption cross-section for thermal neutrons (7.4 barns for 232 Th against 2.7 barns of ^{238}U).

them like ²¹²Bi and ²⁰⁸Tl emits strong γ radiations (0.7-1.8 MeV and 2.6 MeV, respectively). This makes handling very difficult and adds a high degree of self-shielding that can be regarded as protecting the ²³³U from being employed into a weapon. In similar way, ²³²U isotope could be utilized as an attractive carrier of highly enriched uranium (HEU) and weapon grade plutonium (WG-Pu) to avoid their use for non-peaceful purposes. From these considerations, it is clear that incineration of WG-Pu or civilian plutonium in "once-through" cycle based on (Th, Pu)O₂ fuel is more attractive, as compared the (U, Pu)O₂ fuel cycle, because plutonium is not bred in the Th-MOX fuel and ²³²U formed in the spent fuel provides a degree of selfprotection against the risk of proliferation.

From the technological point of view the main challenges associated to the use of Th-MOX fuels are related to the higher sintering temperature required to produce "high density" ThO₂-based mixed oxide fuels, to the need of remote, automated and heavily shielded hot cell for reprocessing and re-fabrication and to the long cooling time required (at least one year) for completing the decay of ²³³Pa to ²³³U [7]. Finally, it should be reminded the potential impact of Th-MOX fuels on the key operational safety parameters due to reduced value of the effective delayed neutron fraction (β_{eff}).

NEUTRONIC PERFORMANCE OF MOX AND TH-MOX FUELS

In order to study the potential use of a fuel cycle based on Th-MOX fuel, some preliminary lattice calculations based on a typical 17x17 PWR fuel assembly design, have been performed. The investigation was aimed to compare the neutronic performance of MOX and Th-MOX fuels in terms of reactivity, impact of burnuble poison and isotopic evolution at different burnup levels. A specific lattice code have been used for this scope. Main results of this analysis are reported in Figures 1 and 2.

In the Figure 1 trend of k-infinity vs. burnup for a MOX fuel (91.8 wt% of 238 U and 8.2 wt% of fissile plutonium i.e. 239 Pu + 241 Pu), and of a Th-MOX, fuel (91.4 wt% 232 Th and 8.6 wt% of fissile plutonium), are compared. As shown in the figure, Th-MOX fuel provides a curve of k-inf vs. burnup quite similar to that of the MOX fuel; also the effect of burnable poison (each fuel assembly includes 20 pins enriched with 4 wt% of gadolinium (Gd₂O₃) to compensate the reactivity excess at beginning of life (BOL)) is comparable in both fuels.

In Figure 2, the evolution of total amount of fissile plutonium versus burn-up for MOX and Th-MOX fuels, is shown; in the same figure the amount of 233 U isotope produced by conversion of 232 Th, is also reported.

As clearly shown in the Figure 2, Th-MOX fuel behaves as more efficient "plutonium burner" if compared with traditional MOX fuel, thanks to the fact that plutonium is not produced in the fuel as consequence of a transmutation process involving the 238 U isotope.



Figure 1 – Trends of k-infinity vs. burnup for MOX and Th-MOX fuels.



Figure 2 – Evolution of fissile plutonium vs. burnup for MOX and Th-MOX fuels (in dotted red-line the evolution of 233 U isotope).

It is also important to stress that significant amounts of the 233 U isotope are accumulated in Th-MOX fuel at high burnup level only (below 40 GWd/t, less of 1 wt% of 233 U is accumulated in the fuel).

Finally, comparable values of the effective delayed neutron fraction (β_{eff}) are obtained for both fuels (365 pcm for MOX fuel, 325 pcm for Th-MOX fuel).

REFERENCE CORE DESIGN

The objective of the present work is to verify the sustainability of a fuel cycle based on thorium-plutonium mixed oxides in a Small Modular Reactor. For that reason, the performances of a core based on Th-MOX fuels will be compared with those of a "reference core design" that use tradition MOX fuels.

The reference SMR core is that of a typical 150 MW thermal power pressurized water reactor designed for being operating with MOX 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 according to the scheme of Figure 1 (first core loading).



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

Fig.1. Core loading pattern at beginning of life.

Each assembly (pattern 17x17), contains 264 fuel rods and has a mechanical and geometrical designs typical for a large PWRs (see Figure 2). The power density in the core is assumed to be 100 kW/l while the active length is 1.35m. The fuel rods are made of Zircaloy tubing containing MOX fuels, a mix of plutonium and ²³⁸U in different wt% depending on their locations in the core (see Table 1). The plutonium vector considered at BOL is reported in Table 2; it is representative of the composition of a typical spent PWR fuel (4.2 wt% ²³⁵U initial enrichment, 50 GWd/t discharge burnup, 10 years of cooling) [2]. Accumulation of ²⁴¹Am from decay of ²⁴¹Pu was neglected so that ²⁴¹Am does not appear in the Pu vector.

It should be pointed out that the maximum amount of plutonium has been limited to 14 wt% for safety reasons in order to avoid any risk of positive reactivity void coefficient in case of generation of coolant voids [8].

Table 1. Plutonium and uranium content in the MOX fuel.

Cycle #	F/A type ^(*)	Pu ^{fiss} (wt%)	Pu ^{tot} (wt%)	²³⁸ U (wt%)
	A11	8.2	12.3	87.7
1	B11	6.3	9.4	90.6
>1	A11	8.2	12.3	87.7

(*) See Fig. 1 for the location of F/A in the core.

Table 2. Abundance of plutonium isotopes in fresh MOX fuel (wt%).

²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu
2.5	54.2	24	12.6	6.7

The assemblies contain fuel rods with gadolinium (Gd_2O_3) as burnable poison to compensate the reactivity excess at beginning of life (BOL). The core has a shutdown system made of 12 Rod Cluster Control Assemblies (RCCAs) each of them containing 24 absorber rods (Ag, In, Cd) over a length which covers nearly the complete active fuel length (see Figure 2).



Fig. 2. Assembly fuel pattern.

The core is axially and radially surrounded by a layer of reflector assemblies of structural materials (stainless steel, Zircaloy) and filled with water. Diluted control material (natural boron) in the coolant is used to control slow reactivity changes during power operation (Xe-poisoning and burn-up effects) and to compensate large reactivity changes during cool-down or heat-up phases. The main core and assembly data are reported in Table 3 with related references. 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 8.2 wt% of fissile plutonium.

Table 3. Core and assembly design data.

Parameter	Unit	Value	Remarks
Total N° F/A.	-	24	Ref. 6
Total N° fuel rods	-	6336	Ref. 6
Fuel pattern	-	17 x 17	Ref. 6
Fuel assembly pitch	cm	21.5	Ref. 5
Pin pitch	cm	1.26	Ref. 5
N° fuel pins per ass.	-	264	Ref. 6
Contr. rod pins per ass.	-	24	Ref. 5
N° instr. Tube per ass.	-	1	Ref. 5
Active length	cm	135	Ref. 6
Fuel pellet diameter	cm	0.819	Ref. 5
Cladding material	-	Zr-4	Ref. 6
Control rod-instr. data	-	-	Ref. 5
Enrich. Pu ^{fiss} @ BOL	wt%	8.2 - 6.3	Assumed
N° fuel zones @ BOL	-	2	Assumed

Total fuel volume	m ³	1.50	-
Axial/radial refl. data	-	-	Ref. 5
Density power	kW/l	100	Assumed
Total thermal power	MWth	150	-
Coolant flow rate	kg/s	721	(*)
Core pressure	MPa	15.5	Ref. 6
Rated coolant flow	kg/cm ² h	234	Calculated
Inlet coolant temper.	°C	295	Ref. 6
Outlet coolant temper.	°C	330	Ref. 6
Average fuel temper.	°C	622	Ref. 6
Ave. moderator temper.	°C	313	Ref. 6
Gd_2O_3 pins per ass.	-	20-16	Assumed
Gd ₂ O ₃ enrichment	wt%	4	Assumed
RCCAs (Ag, In, Cd)	wt%	80, 15, 5	Ref. 6

(*) Scaled from data of large PWR core.

PERFORMANCE OF THE REFERENCE CORE

The analysis of the core behavior in steady state conditions up to the equilibrium cycle has been carried out by using ad-hoc lattice code and 3-D core simulator. A summary of the main core physics characteristics is reported in Table 4. Figures 3 shows the trend of critical boron versus burnup, while in the Figure 4 the axial averaged power shapes at BOC and EOC, are reported. As it can be seen, reasonable values for main core parameters including reactivity coefficients (boron, Doppler and moderator temperature coefficients), are obtained (all negative). However, lower values of the critical boron in the core would be expected by the use of enriched boron in the coolant (> 20% of enrichment in the isotope ${}^{10}B$)². As far as the isotopic evolution, Table 5 reports the plutonium incineration performance; the amount of fissile plutonium reduces of 9.4% (-1.27 kg/assembly) corresponding to an average consumption in the core of around 2.2 kg Pu^{fiss}/GWd/t.

Table 4. Reference core: main core parameters at equilibrium cycle.

Parameter	Unit	Value
Critical boron (HFP,ARO, BOC)	ppm ¹⁰ B	2221
Cycle length	GWd/t	13.60
Max. ass-2D relative power	-	1.34
Max. assembly burnup (EOC)	GWd/t	30.17
Boron coefficient (HFP, BOC)	pcm/ppm	-1.92
Doppler (HFP, BOC)	pcm/°F	-1.60
MTC (HFP, BOC)	pcm/°F	-22.3
$\beta_{\rm eff}$ (BOC)	pcm	370

Legend: HFP = Hot Full Power, ARO = All RCCAs Out, MTC = moderator temperature coefficient.



Fig. 3 – Reference core: boron concentration (in ppm) versus burnup at equilibrium cycle.



Fig. 4 – Reference core: axial averaged power shapes at BOC, and EOC.

Table 5 – Reference core: plutonium incineration performance at equilibrium cycle.

Parameter	Unit	Value
Initial Pu	kg/ass.	20.4
Discharged Pu	kg/ass.	19.1
% Pu burnt		-6.4%
Initial fissile Pu	kg/ass.	13.48
Discharged fissile Pu	kg/ass.	12.21
% Fissile Pu burnt		-9.4%

CORE DESIGN BASED ON TH-MOX FUEL

The starting point is the reference core design with MOX fuels replaced by new fuel assemblies containing a mix of plutonium and ²³²Th, according to the loading scheme of Table 6. The plutonium vector considered at BOL is that already assumed for the reference core and reported in Table 2. Contents of fissile plutonium in the Th-MOX fuels (8.7 wt% for A11 F/As type and 6.7 wt% for B11 F/As type) have been selected with the aim to assure trends of the k-infinity vs. burnup, as much close as possible to those shown by MOX fuels (see Figure 1). Mechanical and geometrical characteristics

² A more negative value of the boron coefficient is also expected.

of the F/As as those of the reference core while no modifications on number of pins and content of burnable poison, have been considered. Thermal-hydraulic operating conditions, rated power, and reactivity control systems are those already considered for the analysis of the reference core.

Table	6.	Plutonium	and	thorium	content in	the	Th-MOX fuel.
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Cycle	F/A	Pu ^{fiss}	Pu ^{tot}	²³² Th
#	type ^(*)	(wt%)	(wt%)	(wt%)
	A11	8.6	12.84	87.16
1	B11	6.7	10	90.0
> 1	A11	8.6	12.84	87.16

(*) See Fig. 1 for the location of F/A in the core.

PERFORMANCE OF TH-MOX FUELED CORE

The analysis of the core behavior in steady state conditions up to the equilibrium cycle has been performed by the same lattice code and 3-D core simulator previously used. Main core physics characteristics are reported in Table 7, in Figure 6 (trend of critical boron versus burnup) and Figure 7 (axial averaged power shapes). As it can be seen Th-MOX fueled core requires, in comparison with reference core, a lower boron concentration in the moderator at BOC (-429 ppm ¹⁰B), has higher cycle length (+0.54 GWd/), provides a higher fuel burnup at discharge (+1.45 GWd/t) while no significant modifications are obtained for the axial power distributions, at different burnup levels, and in the values of reactivity coefficients.

Table 7. Th-MOX core: main core parameters at equilibrium cycle.

Parameter	Unit	Value
Critical boron (HFP,ARO, BOC)	ppm ¹⁰ B	1792
Cycle length	GWd/t	14.14
Max. ass-2D relative power	-	1.36
Max. assembly burnup (EOC)	GWd/t	31.62
Boron coefficient (HFP, BOC)	pcm/ppm	-2.12
Doppler (HFP, BOC)	pcm/°F	-1.83
MTC (HFP, BOC)	pcm/°F	-26.2
$\beta_{\rm eff}(\rm BOC)$	pcm	334

Legend: HFP = Hot Full Power, ARO = All RCCAs Out, MTC = moderator temperature coefficient.



Fig. 6 – Th-MOX core: boron concentration (in ppm) versus burnup at equilibrium cycle (in dotted blue-line the curve for reference core).



Fig. 7 - Th-MOX core: axial averaged power shapes at BOC and EOC (in green/brown markers the results for the reference core).

As far as the isotopic evolution, Table 8 reports the plutonium incineration performance at the equilibrium cycle while in Figure 8 the evolution of fissile plutonium (in wt%) is also shown. The amount of fissile plutonium reduces of 17.4% (-2.17 kg/assembly) corresponding to an average consumption in the core of around 3.7 kg Pu^{fiss}/GWd/t.

From these results it is clear that the SMR based on Th-MOX core is almost double more "efficient" in the burning of fissile plutonium in comparison with the reference core, while preserving reactor operability (cycle length, maximum fuel burnup at discharge) and safety characteristics (negative reactivity coefficients). Moreover, the higher "Pu-burning" characteristics of Th-MOX fuel also contributes to the minimization of the radiotoxicity of spent fuel with clear benefits for the back-end fuel management.

Table 8 – Th-MOX	core: plutonium	incineration	performance at
equilibrium cycle.			

Parameter	Unit	Value
Initial Pu	kg/ass.	19.0
Discharged Pu	kg/ass.	16.7
% Pu burnt		-12.1%
Initial fissile Pu	kg/ass.	12.45
Discharged fissile Pu	kg/ass.	10.28
% Fissile Pu burnt		-17.4%



Fig. 8 - Th-MOX core: isotopic evolution of fissile plutonium at equilibrium cycle (in dotted blue-line the curve for reference core).

CONCLUSIONS

The use of thorium in the form of mixed oxide with plutonium as nuclear fuel, can results in several benefits for the reduction of civil and weapon grade plutonium stockpiles. In view of a possible diffusion of a nuclear fleet of Small Modular Reactors, the feasibility of a fuel cycle based on Th-MOX fuels, has been analyzed in the present work. The SMR core based on Th-MOX fuels has proven to be almost double more "efficient" in the burning of fissile plutonium in comparison with the analogous SMR core loaded with MOX fuels, while preserving the reactor operability and safety characteristics. However, higher amount of fissile plutonium could be incinerated by the optimization of the core design so to achieve higher fuel burnup at discharge (nevertheless significant amount of ²³³U would be expected in the core).

Finally, the absence of new plutonium accumulated in the core through the conversion of fertile isotopes, as well as the radiological barrier introduced by the ²³³U isotope, would assure to the Th-MOX fuels, a high degree of inherent proliferation-resistance against any possible misuse of nuclear material for non peaceful application.

NOMENCLATURE

ARO	All RCCAs Out
BOC	Beginning-Of-Cycle
BOL	Beginning-Of-Life
EOC	End-Of-Cycle
F/A	Fuel Assembly
HEU	Highly Enriched Uranium
HFP	Hot Full Power
LWR	Light Water Reactor
MOX	Uranium-plutonium mixed oxide fuel
MTC	Moderator Temperature Coefficient
RCCA	Rod Cluster Control Assembly
SMR	Small Modular Reactor
Th-MOX	Thorium-plutonium mixed oxide fuel
WG-Pu	Weapon Grade Plutonium

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