Promises and Challenges of Thorium Implementation for Transuranic Transmutation - 13550

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ABSTRACT

This paper focuses on the challenges of implementing a thorium fuel cycle for recycle and transmutation of long-lived actinide components from used nuclear fuel. A multi-stage reactor system is proposed; the first stage consists of current UO_2 once-through LWRs supplying transuranic isotopes that are continuously recycled and burned in second stage reactors in either a uranium (U) or thorium (Th) carrier. The second stage reactors considered for the analysis are Reduced Moderation Pressurized Water Reactors (RMPWRs), reconfigured from current PWR core designs, and Fast Reactors (FRs) with a burner core design. While both RMPWRs and FRs can in principle be employed, each reactor and associated technology has pros and cons. FRs have unmatched flexibility and transmutation efficiency. RMPWRs have higher fuel manufacturing and reprocessing requirements, but may represent a cheaper solution and the opportunity for a shorter time to licensing and deployment. All options require substantial developments in manufacturing, due to the high radiation field, and reprocessing, due to the very high actinide recovery ratio to elicit the claimed radiotoxicity reduction. Th reduces the number of transmutation reactors, and is required to enable a viable RMPWR design, but presents additional challenges on manufacturing and reprocessing. The tradeoff between the various options does not make the choice obvious. Moreover, without an overarching supporting policy in place, the costly and challenging technologies required inherently discourage industrialization of any transmutation scheme, regardless of the adoption of U or Th.

Key Words: transmutation, thorium, reduced moderation, fast reactors

INTRODUCTION

While not eliminating the need for a permanent disposal site, transmutation of long-lived actinides can in principle offer significant benefits from both societal and technical standpoints. It can mitigate public concerns by drastically reducing the amount of long-lived components to be segregated from the biosphere and it can improve repository performance by reducing the heat load of the waste packages during long term disposal. In addition, by recovering and recycling actinides from used fuel, implementation of a transmutation strategy can greatly enhance use of nuclear resources, which can be of strategic importance if nuclear energy is continued in the future. On the other hand, transmutation adds significant costs, risks and technical challenges to the nuclear fuel cycle; it requires large investments and long-term commitment; it can pose safety hazards through handling and manufacturing fuel with isotopes with high specific radioactivity and decay heat. This paper discusses some of the challenges of actinide recycle and

transmutation by analyzing a multi-stage transmutation scenario, starting with an interim Pu burn in current LWRs and implementing full actinide recycle in advanced reactors, specifically Reduced Moderation PWRs and Fast Reactors. The performance with either U or Th-based fuel is analyzed to show potential pros and cons of each option, the areas deserving further investigation and the needed future developmental programs.

TRANSMUTATION SCHEME

The primary goal of the transmutation scheme evaluated in this paper is recycling and burning long-lived transuranic isotopes (TRU) employing the minimum number of transmutation reactors to reduce associated costs. A multi-stage reactor scheme is employed as depicted in Fig. 1. The reactors in the first tier are the current LWRs, currently the large majority of the US and worldwide nuclear fleet, projected to be so for the current century due to their industrial maturity and forecasted new builds. These reactors are assumed to operate with the current fuel cycle and infrastructure to attain optimal economic performance. A second-tier of reactors is then assumed to be gradually introduced for burning the actinides recovered from the used fuel of the first-tier reactors in either a U or Th fertile carrier. As the TRU loading supplied to these reactors will not be completely burned in a single irradiation cycle, and newly generated long-lived actinides will be produced, these reactors need to enable continuous recycle of actinides in their own discharged fuel, for an indefinite number of irradiation cycles. By pursuing full actinide recycle, the High Level Waste (HLW) generated by this nuclear system will in principle consist of fission products (FPs) plus some inevitable but low levels of actinide losses.

The reactors envisaged for the second tier are: Reduced Moderation Pressurized Water Reactors (RMPWRs), and Fast Reactors (FRs). The harder neutron spectrum of RMPWRs, and especially FRs, increases the likelihood of TRU destruction by fission instead of transmutation to higher mass number which is advantageous to implement a continuous recycle policy. While both RMPWRs and FRs can in principle accomplish continuous actinide recycle, FRs offer superior fuel cycle performance and flexibility. They can attain higher thermal efficiency and can be designed as breeders if so desired. By virtue of the higher discharge burn-up (BU), FRs enable significantly lower fuel reprocessing and manufacturing requirements than the counterpart RMPWRs. On the other hand, as evolution of current PWRs, RMPWRs have potentially shorter time to licensing and deployment than FRs, plus some reactors in the current US reactor fleet could be converted to the reduced moderation mode of operation. This may reduce and delay the investment required for building FRs. The technology and infrastructure to support either reactor types requires a certain degree of development, lead time, financial risks and probable economic penalty compared to the current cycle.

A detailed description of the irradiation scheme can be found in [1] and is only summarized here. As mentioned, TRUs recovered via reprocessing of UO_2 LWR discharged fuel are recycled and burned in RMPWRs or FRs. As TRU are irradiated within the Th or U fertile matrix, they are only incompletely burned over a single irradiation cycle with a significant residual fraction, including a portion transmuted to higher actinides (Am, Cm etc.), to be recycled and burned in subsequent irradiation cycles. U, primarily U-233 (“U3” for brevity) and Pa are also generated in Th, which compared to the U scheme entails an additional set of isotopes to be recovered via reprocessing to pursue the full actinide recycle policy. Th fuel also has a lower technology

readiness level compared to U fuel and it introduces additional challenges. However it has specific benefits for TRU transmutation and recycle that may make it worth considering. While the absence of the conversion of U-238 to Pu-239 favors higher Pu destruction, once the fuel is irradiated the U3 produced will reduce the fissile Pu consumption rate. Exchanging Pu with U3 in the fuel inventory, as well as employing natural Th instead of U as the fertile carrier, is however beneficial to mitigate positive reactivity insertion upon core voiding, a known issue of TRU multi-recycle. This advantage is likely essential to the viability of a TRU-burner RMPWR, while it is desirable, but not crucial, for a FR burner. Th-based fuels have also better thermo-physical properties than the U-based counterparts (higher melting point and thermal conductivity) which together with some favorable, but limited, irradiation experience, lead to positive expectations for fuel performance [2].

In addition to direct transmutation of TRU in the second tier reactors, the impact of gaining an interim reduction of Pu by using it in U-PuO_2 (MOX) or Th-PuO_2 (ThOX) fuel for PWRs has also been considered, an option that is shown in the bottom-left part of Fig. 1. In fact, ThOX could be the more credible short-term pathway for Th introduction in the fuel cycle when burning Pu is the primary objective. It still requires some infrastructure development but has a higher consumption of Pu and more stable fuel matrix than comparable U fuel, which may make it an attractive once-through Pu disposition option without precluding a future policy of full recycle. The main advantage of the intermediate ThOX or MOX burn then would be an upfront reduction of Pu in reactors already available, which would minimize the burden of transmutation reactors, RMPWRs and FRs, in the next stage. The drawback, especially notable for FRs, is a larger proportion of higher actinides in the second-stage reactors fuel inventory compared with direct transmutation of TRUs from UO_2 LWRs.

In the scheme investigated, after completing one irradiation cycle in PWRs, used MOX or ThOX fuel would be cooled, reprocessed, with FPs buried as waste and actinides, minus losses, recovered and recycled in the second-tier reactors: RMPWRs and/or FRs. The Np, Am and Cm separated from the original LWR fuel that provided the Pu for ThOX or MOX fuel would be in the second-tier reactors, which is the option considered in the set of calculations with results to follow. Since U3 and Pa, in addition to TRU, will be recovered and recycled in the Th-based transmutation schemes, their consumption is credited as part of the incineration rates reported later on.

At the end of the irradiation cycle, the fuel discharged from the RMPWRs/FRs is cooled and reprocessed. Actinides minus the losses are recycled and remanufactured into new fuel for further irradiation cycles. Only FPs and actinide losses are buried as waste thus in theory achieving a dramatic reduction in the amount of long-lived actinides to be permanently disposed, which is the main objective, and claimed advantage, of continuous recycling schemes compared to once-through operation with direct fuel burial or partial actinide recycle.

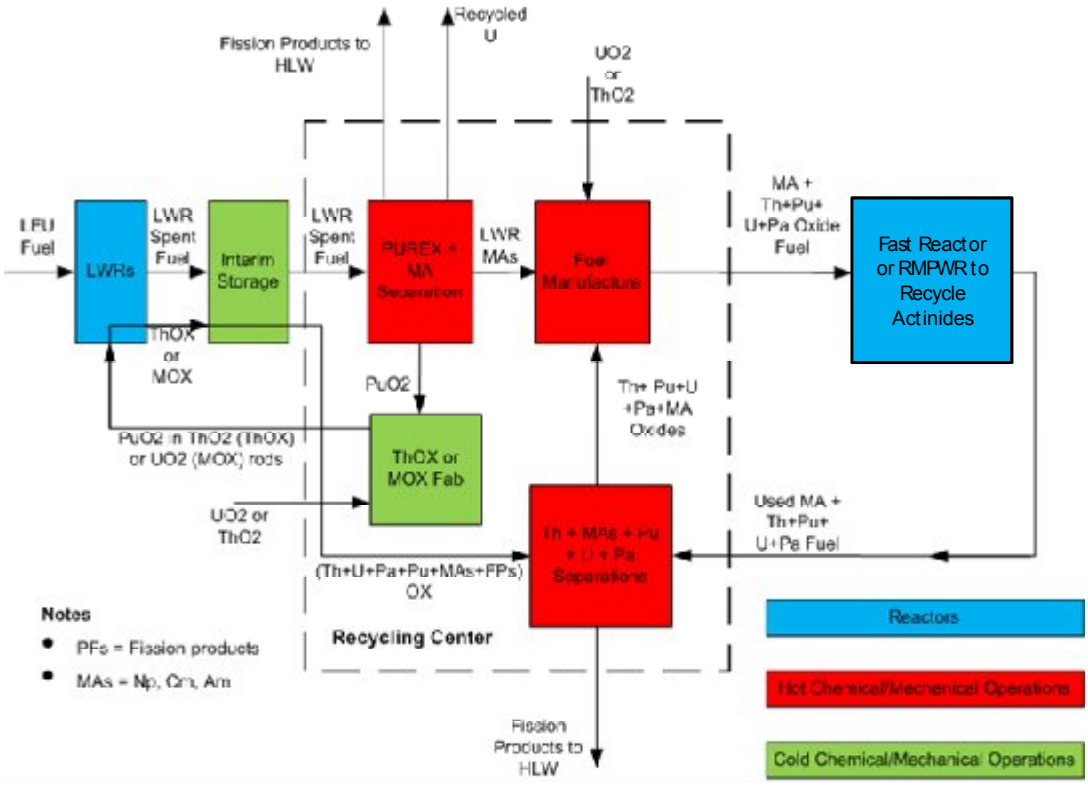

Figure 1 –Fuel Cycle Scheme Analyzed with U-or Th-based Actinide Recycle

RESULTS

A top-level quantitative analysis of the scheme in Fig. 1 has been performed adopting representative reactor designs for the reactor physics analysis, with main features described below.

- Tier 1: 4-loop PWR of Westinghouse design with 3.4 GWt operating on UO_2 fuel with 4.4 % U-235 enrichment, discharge burnup (BU) of 52 GWd/tiHM and 10-yr cooling before recycle of TRU (or Pu) in the next stage(s). When the Pu burn in MOX or ThOX fuel is adopted, the same reactor design and power rating has been assumed with a discharge BU of 50 GWd/tiHM. A cooling time of 5-yr for MOX or ThOX has been assumed before recycle in Tier 2 RMPWRs or FRs.

- Tier 2: Reduced Moderation PWR, based on 4-loop PWR in 1) and 1a) but with larger fuel rod bringing the H/HM from ~2.0 of standard moderation designs to 1.1. This option has been examined only with Th as fertile material. The FR is based on the Westinghouse-Toshiba Na-cooled 1 GWt Advanced Recycling Reactor, ARR [3] with the same recycling schemes as the RMPWR (e.g., direct recycle of TRU from UO_2 PWR fuel or from the interim MOX or ThOX stage). This option has been examined with both U and Th as fertile materials.

Transmutation Performance

Table I summarizes the main results obtained for the fuel cycle scheme depicted in Fig.1, with a focus on the transmutation performance of the various cases. The top row describes fuel and reactor types at the various scenario stages. UO_2 PWR is the first stage ("Stage 1"), common to all scenarios, followed by the optional Pu burn in either MOX or ThOX in a PWR, indicated as "Stage 1a", and then full recycle in the "Stage 2" reactors, i.e., the ARR FR or the RMPWR. When the Pu recovered from Stage 1 is recycled in Stage 1a, the residual Np, Am and Cm from the UO_2 separation are recovered and recycled in Stage 2 together with the TRU (and U3 and Pa for ThOX) from the Stage 1a discharge. If Pu recycle in Stage 1a is not pursued, then all the TRU recovered from UO_2 PWR fuel will be directly recycled in Stage 2 (2nd to 4th row in Table I). The results shown in Table I have been calculated at the equilibrium cycles obtained by repeated recycle in Stage 2 reactors until the mass flows and isotopic inventory of each scenario have converged. The ensuing share of thermal energy for the various stages is derived from the reactors support ratio at these conditions, based on 40% energy conversion efficiency for the FR and 33% for the PWR and RMPWR stages.

The results obtained indicate that direct recycle of actinides from Stage 1 to Stage 2 reactors entails ~30% of the total thermal energy produced by the transmutation reactors, with a ~10% decrease if interim Pu burn in Stage 1a is pursued. This confirms that an intermediate MOX or ThOX stage reduces the number of transmutation reactors, which is particularly desirable for FRs due to the anticipated high capital cost and operating expenses, and long time to deployment. Th-based transmutation appears to have greater potential for minimization of the number of transmutation reactors due to their inferior neutron economy compared to the U FR cycle (lower fast-fission contribution from Th-232 compared to U-238, lower average number of fission neutrons from U3 compared to Pu and, as a result of the lower density compared to U-based oxide fuel, higher leakages with reduced internal breeding). [4]

Table I also shows each stage's incineration rate and the average value, obtained using the share of thermal energy of each stage as weight. As a result of a preceding MOX stage the overall rate of incineration of the U-based scheme decreases from 198 to 182 kg/GWt-yr, while the opposite is true for the Th-based schemes, where it increases from 225 to 238 kg/GWt-yr in the Th-FR and 192 to 223 kg/GWt-yr in the RMPWR. This behavior is explained by the relatively low incineration rate of MOX, less than 100 kg/GWt-yr, preceding the U-FR stage, which itself has a ~ 200 kg/GWt-yr incineration rate with Stage 1a feed. Conversely, the higher incineration rates of ThOX, over 180 kg/GWt-yr, and the Th-FR or RMPWR, 258 and 236 kg/GWt-yr respectively with Stage 1a feed, favorably combine to yield the trend observed. It should be noted that to maximize incineration in the last stage, it is desirable to burn Pu in a preceding thermal-spectrum stage (e.g., ThOX PWR) and avoid conversion in a hard spectrum where the higher fission neutron yield of Pu promotes higher internal breeding. Note also that the higher incineration rate of the Th-FR is reflected in the lower thermal output for Stage 2 reactors across the scenarios studied, i.e. 19% vs. 21% for the RMPWR and 23% for the U FR.

The discharge BUs and kg incinerated/MT reprocessed, shown in Table I are calculated as averages over Stage 1a (when present) and 2. The relatively low discharge BU in the RMPWRs scheme, ~ 43 GWd/tiHM, is a consequence of the tradeoff in increasing TRU loading (and

discharge BU) while respecting safety limits (Moderator Temperature Coefficient, MTC, especially) and operational constraints (reactivity control). The discharge BU in FRs is dictated primarily by respecting the limit on the cladding material fast fluence. As a result, FR-based schemes can achieve much higher discharge BU than the RMPWR, resulting in 110-120 GWd/tiHM and 90-96 GWd/tiHM average BUs for respectively the single and multi-stage schemes (the difference originating from the low discharge BU, ~ 50 GWd/tiHM, of the MOX or ThOX stage compared to FR). Therefore FRs offer a more efficient incineration environment, confirmed by the significantly higher ratio of kg Burned/MT Reprocessed displayed in Table I. The slightly higher discharge BU of the Th vs. U FR schemes is due primarily to the lower density of Th-based fuels, while adopting the same fuel management scheme of U-based fuel; the higher discharge BU, combined with the higher incineration rate, results in the larger values of kg burned/MT reprocessed pertaining to the Th-based FR schemes.

Fuel Manufacturing

Fuel manufacturing and reprocessing are the key technological components of a transmutation scheme. The one investigated here assumes oxide fuel with aqueous reprocessing in recognition of the wider experience basis and potential for implementation on an industrial scale.

Fuel incorporating Pu and/or fresh Th needs to be manufactured in a glove-box facility (shown in green in Fig. 1). The fuel for the second-tier reactors however needs to be performed remotely due to the high radiation field due to the presence of Am, Cm and higher actinides, and for Th-based recycled fuel, U-232. Table I shows the breakdown of fuel manufacturing for each stage of the various schemes in terms of MT of fuel per GWe-yr produced. The dominant economic discriminator is the amount of fuel that needs to be fabricated remotely.

FRs fare significantly better than RMPWRs from this standpoint, with about one third of the fuel requiring remote manufacture, assuming a homogeneous recycling strategy. Under this assumption, the multi-tier scheme would be the optimum and the Th FR would be slightly favored compared to the U FR, with 1.9 MT of fuel fabricated remotely, 2.1 MT in glove-box and ~15 MT hands-on for GWe-yr produced by the entire nuclear system.

If a heterogeneous recycle strategy is pursued, then the amount of fuel requiring remote manufacturing could be reduced significantly, especially for the U-based scheme, since Th-based transmutation would require remote manufacturing of U3-bearing fuel, due to its content of U-232, in addition to Am/Cm bearing fuel. Also, for glove-box fuel remanufacturing of Th-Pu fuel, co-precipitation with ~ 20-year interim storage to allow for decay of Th-228, or partition of Pu to reuse it with fresh Th, would be required. It should be remarked also that the partition into various elemental streams (Th, U3, Pu, Am/Cm) would need to be realized by proper reprocessing technology, and whether this can be achieved is unclear at this point and would likely require considerable development.

Table I Summary of Transmutation Performance

Designation[1]	U-FR-SS	Th-FR-SS	RMPWR-SS	U-FR-MS	Th-FR-MS	RMPWR-MS
Fuel & Reactor Stages 1 Stage 1a Stage 2	UO_2 PWR/ (Absent)/ U-FR	UO_2 PWR/ (Absent)/ Th-FR	UO_2 PWR/ (Absent)/ Th-RMPWR	UO_2 PWR/ MOX-PWR/ U-FR	UO_2 PWR/ ThOX-PWR/ Th-FR	UO_2 PWR/ ThOX-PWR/ Th-RMPWR
Fertile in Stage 1a and 2	U-238	Th-232	Th-232	U-238	Th-232	Th-232
Actinides Burned/Source in Stage 1a	Absent	Absent	Absent	Pu in UO_2	Pu in UO_2	Pu in UO_2
Actinides Burned/Source in Stage 2	TRU/UO_2	TRU/UO_2	TRU/UO_2	TRU/MOX Np,Am,Cm/UO_2	TRU,U3/ThOX Np,Am,Cm /UO_2	TRU,U3/ThOX Np,Am,Cm /UO_2
Share of Thermal Output (%)[2] Stage 1a , Stage 2	 Absent 31%	 Absent 29%	 Absent 32%	 11% 23%	 9% 19%	 7% 21%
Incineration Rate (kg/GWt-yr) Stage 1a Stage 2 (Average)[3]	 Absent 198 (198)	 Absent 225 (225)	 Absent 192 (192)	 94 213 (182)	 184 258 (238)	 184 236 (223)
Burn-up (GWd/tiHM)[3]	110	118	43	90	96	43
kg Burned/MT Reprocessed[3]	60	73	23	47	65	26
MT of Fuel per GWe-yr System Stage 1 Stage 1a Stage 2 (Total)	 13.7 Absent 3.6 (17.3)	 14.3 Absent 3.1 (17.4)	 14.5 Absent 8.2 (22.7)	 13.4 2.4 2.7 (18.4)	 14.8 1.9 2.1 (18.7)	 15.3 1.5 5.8 (22.7)

Notes: (1) “SS”: Single-Stage; “MS”: Multi-Stage. 2) An energy conversion efficiency of 0.33 for the PWR and RMPWR and 0.4 for the ARR FR has been assumed. (3) Average Incineration Rate, Burn-up and kg Burned/MT Reprocessed are averaged over stage 1a and 2, using as weights the thermal share for these two stages

Fuel Inventories

The fuel compositions by weight percent of heavy metal constituents for both start-up and equilibrium cores are shown in Table II. The FR cores feature larger fissile proportions due to the combined effect of higher discharge BU, increased leakage and reduced fissile cross-sections. The higher discharge BU is beneficial to lower manufacturing and separation requirements, as discussed.

As the fuel is irradiated through the various cycles, the isotopes reloaded from the previous stage(s) are only partially consumed in a given irradiation cycle, new fissile is bred from fertile elements, some isotopes are transmuted by neutron capture which leads to a buildup of isotopes with higher mass numbers, and some isotopes decay. Eventually an equilibrium is reached where isotopic consumption and decay equates production and injection as external feed, which when achieved by all isotopes leads to a stable isotopic inventory. As the main neutronic properties are conferred by elements whose content is established relatively quickly, reactivity and mass flows take only a few cycles to converge. On the other hand, some isotopes which may be of low significance for reactor physics considerations but are important for their impact on front-end and back-end, e.g. for their high specific radiation field, or the long-term radiological threat they pose, may evolve for a much longer time. Multi-cycle simulations from start-up to the equilibrium cycle have thus been performed so that the buildup of the meaningful fuel cycle isotopes could be captured and compared across the various cases. More specifically, a total period of in-core irradiation of 110 Effective Full Power Years, (EFPY) plus cycle-to-cycle cooling time, reprocessing and reloading has been simulated. With a 5-year cooling time, this corresponds to over 200 years of recycling, at which point the isotopes at the upper end of the transmutation chain (e.g., Cf-252) are also effectively converged.

The evolution with recycle of the main elements in the fuel inventory can be inferred examining Table II. In the Th single-stage reactors, the buildup of U3 from start-up to equilibrium entails a decrease in the Pu inventory, contrasted to the Th multi-stage reactors where the presence of U3 in the external feed from the prior ThOX stage and the poorer fissile quality of the Pu injected results in the opposite trend. In the U-FR cases where Pu is also bred from U, in addition to the external feed, its content at equilibrium is slightly higher than at start-up. The content of Cm (and typically of Am) is increasing for all schemes, following the common prevailing consumption mechanism of Am, i.e. transmutation by neutron capture, which ultimately leads to the generation of Cm. Notably, despite the lower Pu inventory, the Th-FR cases feature higher Am and Cm inventory compared to the U counterpart: this follows from their higher rate of external feed, which itself contains Am and Cm, which leads to the higher accumulation Am/Cm observed before equilibrium is reached. Note also that the higher fertile proportion, and lower incineration rate, of the RMPWR promotes comparatively lower proportions of Am and Cm, despite the higher chance of transmutation vs. fission in the softer spectrum relatively to the FR. Table II shows a total Am+Cm content for the Th-FR vs. RMPWR of ~5 % vs. 3% in the single stage scenario and 9% vs. 5% for the multi-stage scenario. The implications on the neutron source at fabrication are discussed next. Finally, the specific content of U-232 in the Th FR is twice that of the RMPWR, primarily as a result of the larger fuel proportion of U-233, which has some implications on the gamma source at fabrication to be discussed in the following.

Table II Fuel Inventory

Designation	U-FR-SS	Th-FR-SS	RMPWR-SS	U-FR-MS	Th-FR-MS	RMPWR-MS
Specific Power, MWt/MT	98	104	28	98	104	28
TRU+U3 External Feed (kg/GWt-yr)	198	225	192	213	258	236
Fuel Composition, weight % (Start-up Core) Equilibrium Core						
Natural Th or U	(66.9%) 61.9%	(61.2%) 55.1%	(79.5%) 76.2%	(59.0%) 57.5%	(55.6%) 49.1%	(79.5%) 68.4%
U3	-	(-) 8.8%	(-) 5.6%	-	(10.4%) 13.8%	(3.8%) 7.5%
Np	(1.5%) 0.8%	(1.8%) 1.1%	(1.0%) 0.5%	(3.1%) 1.4%	(2.9%) 1.7%	(1.3%) 0.9%
Pu	(29.0%) 33.1%	(34.0%) 29.8%	(17.9%) 14.5%	(31.2%) 34%	(23.9%) 26.2%	(12.0%) 18.1%
Am	(2.4%) 3.1%	(2.8%) 3.8%	(1.5%) 2.2%	(5.6%) 4.8%	(6.0%) 6.1%	(3.0%) 3.7%
Cm	(0.2%) 1.1%	(0.2%) 1.3%	(0.1%) 1.0%	(1.2%) 2.4%	(1.2%) 3%	(0.4%) 1.4%
U-232 (ppm of HM)	0	409	223	0	543	275

Neutron Field

As discussed, regardless of the spectrum, isotopic multi-recycle inevitably leads to some accumulation of higher actinides, Cm and eventually Cf, especially when some of these isotopes (Cm) and their precursors (Am) are also supplied as external feed. As a result, a considerable neutron source, predominantly from spontaneous fission reactions, arises. The neutron source at fabrication for the various schemes is shown in Table III. The table also recalls the fuel requirements of the second stage reactors, expressed as MT of HM and Number of Assemblies per GWe-yr produced by the entire system (including the pertaining prior stages). This is an important discriminator for the various options, as remote fuel manufacturing requirements are expected to be among the dominant economic and technological factors.

The sources given in Table III are expressed as both n/s/gHM as well as n/s/assembly, as either choice may be better suited to confer the implications at different manufacturing stages, or at fuel handling and transportation. The source is shown at four cumulative irradiation times to show the impact of recycling: start-up core ("0 EFPY"), 21 EFPY (~50 years worth of recycling, including cooling times), 42 EFPY (~100 years of recycling) and 110 EFPY, which can be considered as an asymptotic value. The source has been evaluated supposing 5 years of decay from reactor discharge to the next round of fabrication,

accounting for cooling, reprocessing and top-up of recycled fuel with external feed. The bottom part of Table III shows the main contributors to the neutron source.

All cases show a considerable neutron source, in the range of 10^4 to 10^6 n/s gHM, which translates into 10^8-10^{12} n/s/assembly. These sources require significant shielding, on the order of several feet of concrete, to maintain radiation exposure to the workers within acceptable limits. The RMPWR tends to have higher specific source due to larger generation of higher actinides in the softer spectrum; this impacts particularly the generation of californium (Cf), since the content of Am and Cm is driven primarily by the external feed rate and so these isotopes are present at high content also in the FRs. The higher content of Am/Cm in the external feed to the multi-stage transmutation scheme leads to higher neutron sources, especially during the first recycles. The higher proportion of fertile isotopes of the RMPWR compared to the FR counterbalances the higher generation rate of neutron emitters until the buildup of Cf becomes the predominant factor (after ~20 EFPY for the multi-stage RMPWR scheme and ~30 EFPY for the single-tier scheme). For the FRs, the leading neutron source contributor remains Cm, even though the contribution of Cf increases as the irradiation progresses.

The U and Th FRs have very similar neutron sources, typically slightly higher for Th as a result of the higher feed rate of Am/Cm. There is however a significant difference in the gamma source between U and Th, as will be discussed next.

Table III Neutron Source and Its Components

Designation	U-FR-SS	Th-FR-SS	RMPWR-SS	U-FR-MS	Th-FR-MS	RMPWR-MS
Fuel Requirements:						
MT/GWe-yr System	3.6	3.1	8.2	2.7	2.1	5.8
Assembly/GWe-yr System	139	127	13	104	86	9
n/s/gHM						
0 EFPY	1.6E+04	1.9E+04	1.2E+04	1.2E+05	1.2E+05	3.7E+04
21 EFPY	6.8E+04	7.6E+04	6.5E+04	2.1E+05	2.3E+05	1.4E+05
42 EFPY	1.1E+05	1.1E+05	5.9E+05	2.9E+05	3.1E+05	3.2E+05
110 EFPY	1.6E+05	1.7E+05	1.6E+06	4.2E+05	4.4E+05	2.0E+06
n/s/assembly						
0 EFPY	4.24E+08	4.65E+08	7.70E+09	2.70E+09	2.55E+09	2.33E+10
21 EFPY	1.77E+09	1.85E+09	4.11E+10	4.90E+09	5.14E+09	8.75E+10
42 EFPY	2.75E+09	2.77E+09	3.78E+11	6.68E+09	6.80E+09	2.04E+11
110 EFPY	4.20E+09	4.11E+09	1.04E+12	8.99E+09	9.14E+09	1.27E+12
% Contribution of Cm, (Cf)						
0 EFPY	98% (0%)	98% (0%)	98% (0%)	99% (0%)	99% (0%)	99% (0%)
21 EFPY	96% (3%)	97% (2%)	62% (37%)	94% (5%)	96% (3%)	53% (46%)
42 EFPY	78% (21%)	85% (14%)	16% (84%)	75% (24%)	83% (17%)	18% (82%)
110 EFPY	58% (41%)	65% (34%)	5% (95%)	59% (41%)	67% (33%)	6% (94%)

Gamma Field

The primary gamma source, expressed as MeV/s/gHM and tabulated for various energy bins, is given in Table IV. A plot showing the main contributing isotopes is given in Fig. 1. The secondary gamma source from (n,gamma) reactions in the surrounding material is not considered here, but it represents an additional contributor in the determination of shielding requirements. The low gamma energy components, up to ~200 keV, of the primary gamma source are dominated by the decay of Am-241 and 243, Cm-244 and Pu-238, and so are the same or similar in U and Th fuels. However, Th shows an additional significant gamma field at higher energy which is due to the decay of the U-232's daughters, primarily Bi-212 and Tl-208. Tl-208 in particular is responsible for the emission of an intense and particularly penetrating 2.6 MeV gamma ray. U-232 is an unavoidable by product of the Th cycle, especially in the driver fuel where it is generated by high energy neutron (n,2n) reactions with Th-232 and U-233; it builds up relatively quickly, reaching equilibrium during the first few cycles of irradiation.

The higher content of U-232 in the Th FR fuel inventory (see last row of Table II) leads to a larger gamma field, about twice in the 2.5-3 MeV bin, compared to the RMPWR. Aside from the first cycles, the gamma field does not show large variations and is typically higher for the multi-stage vs. the single-stage recycling options, as an implication of the larger content of U-232, Cm-244 and Am. There is however a marked temporal dependence in the high energy gamma source with the time after reprocessing, when the daughters of U-232 are temporarily eliminated from the recycled fuel inventory as a result of the separation stage, and then resume their growth to secular equilibrium. If Th is partitioned during the reprocessing and not immediately recycled, then U-232's first and longest-lived daughter, the 1.9-year half-life Th-228, would be initially absent and the growth of U-232's daughters would be significantly delayed as now dependent on the 69 year half life U-232. The impact on the buildup of U-232's daughters can be appreciated in Figure 2, showing the gamma field at fabrication with fresh (i.e. no Th-228 initially present) or recycled (Th-228 at secular equilibrium) Th, at various times after reprocessing. The gamma field reaches over 70% of its peak already after one week after the separation when Th is immediately recycled, whereas if Th-228 is initially absent the gamma-field after one month is still less than 3% of the peak value. Since the fabrication process can take months (and years considering the waste processing steps within the fabrication plant), there is likely little practical difference between these two options.

The possibility of independently managing Th from the rest of the recycled actinides relies on devising a suitable reprocessing separation technique, plus the potential benefits of delaying the onset of the gamma field depend on the specific fuel manufacturing technique adopted, and would certainly require collocation of separation and manufacturing facilities. Given the shielding requirements of fuel bearing Am and Cm, the benefits of Th partition materialize only if a heterogeneous recycling scheme with independent management of Pu and U3+Am+Cm is a viable option so that Th-Pu can be manufactured in glove-box with only the remaining Th-U3-Am-Cm fuel manufactured remotely. It should also be remarked in this context that the feasibility of fuel manufacturing with a relatively high content of Am and Cm, especially for the FR multi-stage options examined, and even more so for heterogeneous schemes, needs to be determined.

Historically, the thick shielding required to cope with the high-energy gamma radiation associated to U-232 has been a cogent deterrent to the implementation of the Th cycle. In TRU-burning reactor schemes as the one investigated here, this challenge compounds difficulties related to the presence of a high

neutron source, thus likely further penalizing economics. On the other hand, since substantial remote fuel manufacturing and handling would be required regardless of the choice of U and Th, when adopted in a TRU burning context the Th option appears less economically penalizing than in more traditional breeding scheme, at least as far as fuel manufacturing is concerned.

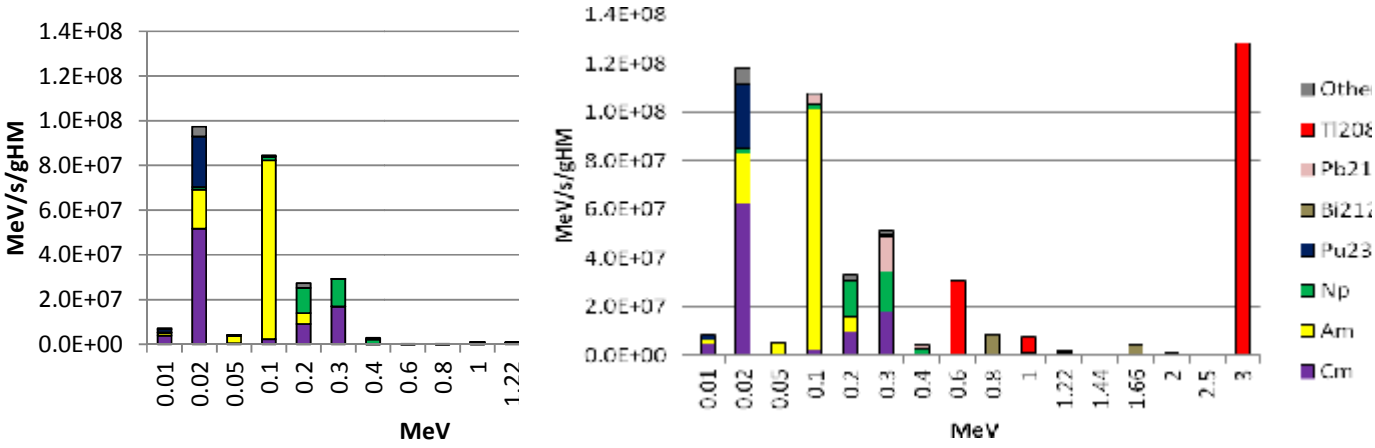

Figure 1 Contributors to Gamma Field at fabrication for the U-FR (left) and Th-FR (right) - 1-year after reprocessing (RMPWR-MS, 5-year cooling before reprocessing, 42 EFPY of prior irradiation)

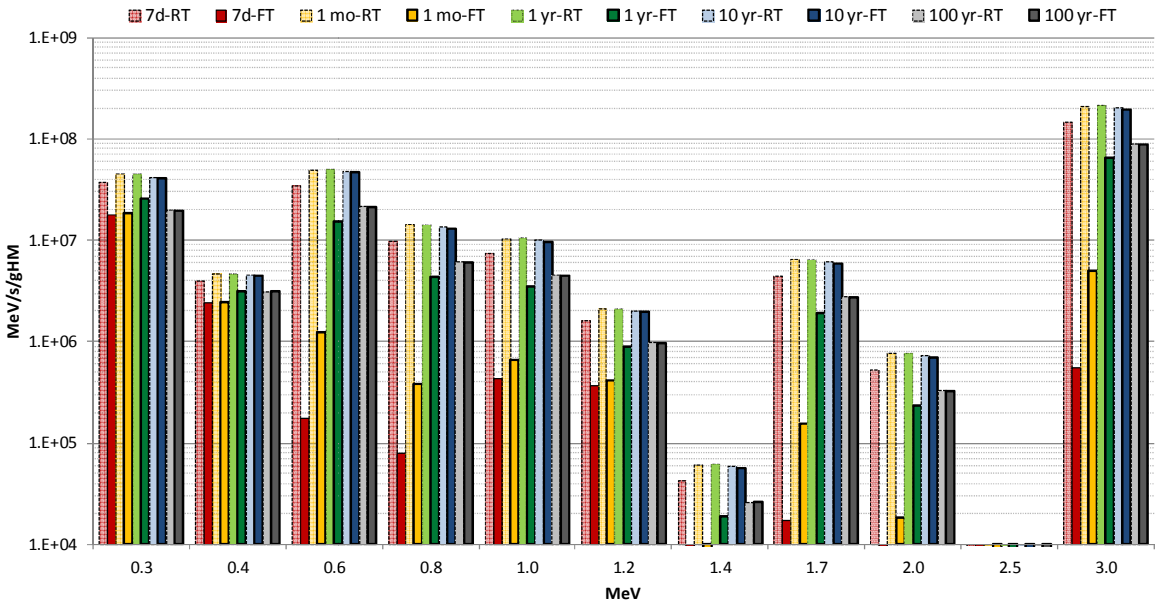

Figure 2 Impact on gamma field of employing recycled ("RT") vs. fresh ("FT") Th (RMPWR-MS, 42 EFPY)
5-year cooling before reprocessing and 7-days, 1-month, 1, 10 and 100 years before fabrication

Table IV Gamma source (MeV/s/gHM) per energy interval (MeV, upper bound) 5-year cooling before reprocessing and 1-year decay before fabrication

MeV/s/gHM / MeV	U-FR-SS	Th-FR-SS	RMPWR-SS	U-FR-MS	Th-FR-MS	RMPWR-MS
0.01	4.0E+06	4.8E+06	3.2E+06	7.1E+06	8.6E+06	4.6E+06
0.02	5.4E+07	6.6E+07	4.5E+07	9.7E+07	1.2E+08	6.5E+07
0.05	2.8E+06	3.5E+06	2.2E+06	4.1E+06	5.1E+06	3.0E+06
0.1	6.0E+07	7.7E+07	4.6E+07	8.5E+07	1.1E+08	6.3E+07
0.2	1.6E+07	1.9E+07	9.5E+06	2.7E+07	3.3E+07	1.4E+07
0.3	1.8E+07	3.3E+07	1.6E+07	2.9E+07	5.1E+07	2.4E+07
0.4	1.3E+06	2.5E+06	1.3E+06	2.8E+06	4.6E+06	2.0E+06
0.6	4.0E+04	2.3E+07	1.1E+07	7.7E+04	3.1E+07	1.6E+07
0.8	3.2E+04	6.5E+06	3.0E+06	5.0E+04	8.6E+06	4.5E+06
1	5.9E+05	5.3E+06	2.4E+06	8.3E+05	7.1E+06	3.5E+06
1.22	5.3E+05	1.4E+06	5.7E+05	7.4E+05	1.9E+06	8.4E+05
1.44	7.5E+00	2.8E+04	1.3E+04	6.5E+00	3.8E+04	2.0E+04
1.66	2.9E+02	2.9E+06	1.3E+06	1.5E+02	3.9E+06	2.0E+06
2	4.7E+01	3.5E+05	1.6E+05	3.0E+01	4.6E+05	2.4E+05
2.5	2.3E-02	1.8E-03	1.3E-03	1.2E-03	3.2E-03	2.0E-03
3	**9.3E+03**	**9.6E+07**	**4.4E+07**	**4.9E+03**	**1.3E+08**	**6.7E+07**
4	1.0E-04	8.2E-06	5.9E-06	2.1E-06	1.5E-05	8.9E-06

Reprocessing

Even assuming that a suitable remote fuel manufacturing technique can be found to produce the industrial amounts of highly radioactive fuel, and an advanced reactor fleet can be deployed for the transmutation, there remain significant technological gaps in the scheme described. A case in point is the extremely high actinide recovery ratio required, exceeding 99.9% for Pu and Am, and including partition of Cm from the FPs, to achieve a HLW with a ~300-year U ore radiotoxicity target. While partition with 99.9% recovery of Pu is conceivable in current state-of-the-art industrial reprocessing, [5] partition with high recovery of Am and Cm require significant development before achieving industrial status. If Th-based transmutation is pursued, recovery and partition of additional actinides is required, e.g. Pa (Pa-231 could be a long-term hazard due to its relative mobility in some repository environments) and the reprocessing itself will be complicated by more difficult dissolution and far less experience than with U fuels. Past experience includes reprocessing of 17 MT of stainless steel clad Th oxide pellet fuel from the Indian Point nuclear plant which was performed successfully at the commercial Nuclear Fuel Services plant at West Valley, NY; previously, over 800 MT of fuel had been reprocessed using the THOREX process at ORNL, SRP and Hanford [6]. However, the composition and BU of the fuel to be recycled now would be substantially different than in the past experience [7], and it is not clear without further investigations and likely a (costly) pilot experimental program what can really be achieved. Reprocessing, together with fuel manufacturing, is certainly one area to be

recommended for further work before discriminating among the various options presented, and assuming a 300-year radiotoxicity as a sensible goal. Reprocessing schemes should also be acknowledged that, while not aiming at a 300-year HLW waste radiotoxicity target, are based on current capabilities and already benefit nuclear fuel cycles and their waste management without requiring the partitioning of Np, Am and Cm from the FPs. For instance reprocessing schemes such as the Thermal Oxide Reprocessing Plant (THORP) [8] at Sellafield in the UK partitions and recovers U and Pu from the rest of the actinides and the FPs. THORP achieves a significant concentration of the radioactive isotopes into a vitrified form more suitable for geologic disposal, and a consequent reduction in high level waste volumes (typically 12 to 40-fold) in comparison with non-reprocessed used fuel [9].

CONCLUSIONS

A multi-tier fuel cycle scheme aimed at devising a credible industrial route for transmutation of long-lived actinides contained in UO_2 used fuel has been described. The first-tier reactors are current LWRs; the second tier reactors recycle and incinerate TRUs supplied from the first tier over multiple, in principle indefinite, in-reactor irradiation cycles. The reactors investigated for this purpose are RMPWRs with Th-based fuel and FRs with either U-based or Th-based fuel. A top-level comparison of the transmutation performance of the various schemes has been provided based on a core physics analysis, together with an assessment of the fuel inventory characterizing each option and the implications on fuel manufacturing and handling.

The results indicate that in principle all options considered can achieve full recycle and incineration of TRU from UO_2 used fuel, which would dramatically reduce the inventory of long-lived isotopes requiring permanent disposal compared to used fuel burial. The Th-based incineration route has greater potential for reducing the number of transmutation reactors, and a minimum is achieved when preceded by an interim Pu burn in Th-PuO_2 PWRs. FRs are more efficient than RMPWRs because even with slightly inferior incineration rates they can attain significantly greater discharge BU, thus lowering reprocessing and fuel manufacturing requirements. U-based transmutation can build on a much more developed knowledge basis and industrial infrastructure and, for heterogeneous recycling schemes, minimizes the amount of fuel requiring remote manufacturing. While part of this infrastructure will not be directly applicable also to U-based transmutation, and substantial further developments will be required anyhow, U appears the more likely short-term option if FRs with heterogeneous transmutation cores are pursued. Th can be appealing if RMPWRs are the preferred option, and has the additional advantage of mitigating reactivity insertion following core voiding with relatively simple fuel design (e.g. axially homogeneous), as discussed in another contribution to this session [10]. On the other hand, the technology gap with respect to U is considerable [11].

Fuel manufacturing in the presence of a severe radiation field is certainly a key challenge of every scheme, particularly of those related to Th where the difficulties of coping with the significant neutron source from Cm and, especially for the RMPWR, Cf, are compounded by the characteristic, and extremely penetrating, gamma rays from the decay products of U-232. Reprocessing is another demanding area for all schemes, mostly because of the high actinide recovery ratio required, including Am and Cm to effect the claimed ~300-year HLW radiotoxicity target. Th is not favored also in this aspect, at least in consideration of the absence of an existing supporting industrial process, due to the increased difficulties in the dissolution stage and the larger span of isotopes to be recovered and recycled.

In general, development of expensive and challenging technologies would be required for all schemes, inherently discouraging industrialization without an overriding, compelling driver.

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