Opportunities for the Multi Recycling of Used MOX Fuel in the US - 12122

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ABSTRACT

Over the last 50 years the US has accumulated an inventory of used nuclear fuel (UNF) in the region of 64,000 metric tons in 2010, and adds an additional 2,200 metric tons each year from the current fleet of 104 Light Water Reactors. This paper considers a fuel cycle option that would be available for a future pilot U.S. recycling plant that could take advantage of the unique opportunities offered by the age and size of the large U.S. UNF inventory.

For the purpose of this scenario, recycling of UNF must use the available reactor infrastructure, currently LWR's, and the main product of recycling is considered to be plutonium (Pu), recycled into MOX fuel for use in these reactors. Use of MOX fuels must provide the service (burn-up) expected by the reactor operator, with the required level of safety. To do so, the fissile material concentration (Pu-239, Pu-241) in the MOX must be high enough to maintain criticality, while, in current recycle facilities, the Pu-238 content has to be kept low enough to prevent excessive heat load, neutron emission, and neutron capture during recycle operations.

In most countries, used MOX fuel (MOX UNF) is typically stored after one irradiation in an LWR, pending the development of the GEN IV reactors, since it is considered difficult to directly reuse the recycled MOX fuel in LWRs due to the degraded Pu fissile isotopic composition. In the US, it is possible to blend MOX UNF with LEUOx UNF from the large inventory, using the oldest UNF first. Blending at the ratio of about one MOX UNF assembly with 15 LEUOx UNF assemblies, would achieve a fissile plutonium concentration sufficient for reirradiation in new MOX fuel. The Pu-238 yield in the new fuel will be sufficiently low to meet current fuel fabrication standards. Therefore, it should be possible in the context of the US, for discharged MOX fuel to be recycled back into LWR's, using only technologies already industrially deployed worldwide. Building on that possibility, two scenarios are assessed where current US inventory is treated; Pu recycled in LWR MOX fuels, and used MOX fuels themselves are treated in a continuous partitioning-transmutation mode (case 2a) or until the whole current UNF inventory (64,000 MT in 2010) has been treated followed by disposal of the MOX UNF to a geologic repository (case 2b). In the recycling scenario, two cases (2a and 2b) are considered. Benefits achieved are compared with the once through scenario (case 1) where UNF in the current US inventory are disposed directly to a geologic repository. For each scenario, the heat load and radioactivity of the high activity wastes disposed to a geologic repository are calculated and the savings in natural resources quantified, and compared with the once-through fuel cycle.

Assuming an initial pilot recycling facility with a capacity of 800 metric tons a year of heavy metal begins operation in 2030, ~8 metric tons per year of Pu is recovered from the LEUOx UNF inventory, and is used to produce fresh MOX fuels. At a later time, additional treatment and recycling capacities are assumed to begin operation, to accommodate blending and

recycling of used MOX Pu, up to 2,400 MT/yr treatment capacity to enable processing UNF slightly faster than the rate of generation.

Results of this scenario analysis study show the flexibility of the recycling scenarios so that Pu is managed in a way that avoids accumulating used MOX fuels. If at some future date, the decision is made to dispose of the MOX UNF to a geologic repository (case 2b), the scenario is neutral to final repository heat load in comparison to the direct disposal of all UNF (case 1), while diminishing use of natural uranium, enrichment, UNF accumulation, and the volume of HLW. Further recycling of Pu at the end of the scenario (case 2a) would exhibit further benefits. As expected, Pu-241 and Am-241 are the source of long term HLW heat load and Am-241 and Np-237 are the source of long term radiotoxicity. When advanced technology is available, introduction of minor actinide recycling, in addition to Pu recycling, by the end of this scenario, or sooner, would have a major impact on final repository heat load and long term radiotoxicity of the HLW. This scenario is also compatible with a gradual introduction of a small number of FR's for Pu management.

INTRODUCTION

Over the last 50 years, the US has accumulated an inventory of used nuclear fuel (UNF) in the region of 64,000 metric tons in 2010, and adds an additional 2,200 metric tons each year. UNF is currently stored in a combination of dry and wet storage at the reactor sites and a few non-reactor sites. A future recycling facility in the US could recycle the U and Pu components (major energy resources) into the existing LWR reactor fleet in the US. AREVA proposes that, as a first step for recycling in the US, a pilot scale 800 tHM/y recycling facility should be designed and built using the best available commercial technology to process LEUOx UNF and recycle the Pu component in LWR MOX.

Any use of MOX fuels must provide the burn-up expected by the reactor operator, with the required level of safety. To do so, the fissile material concentration (Pu-239, Pu-241) in the MOX must be high enough to maintain criticality, while Pu-238 content has to be kept low enough to prevent excessive heat load, neutron emission, and neutron capture during recycle operations. In most countries that use MOX, used MOX fuel (MOX UNF) is typically stored after one irradiation in an LWR, pending the development of the GEN IV reactors, since it is considered difficult to reuse the recycled MOX fuel in LWRs due to the degraded Pu fissile isotopic composition.

The importance of decay time on HLW disposal has been discussed in previous papers [1]. In the US, a large inventory of long decayed UNF has accumulated and presents a unique opportunity for the multiple recycle of MOX UNF by blending with LEUOx UNF, to maintain the fissile Pu isotopic concentration within LWR requirements.

The analyses described in this paper are based on specific US inventory of UNF and describes, a method to meet both the minimum fissile requirement and current fuel fabrication maximum limit for Pu-238 concentration in MOX fuel. As proposed by AREVA, the study assumes recycling occurs initially in an 800 tHM/y Pilot Recycling Facility and allows for future capacity increases needed to procure adequate used MOX blending and to process UNF at the rate of generation, approximately 2400 metric tons per year. The objective of this scenario was to quantify the benefits of a recycling scenario, using LWR MOX to recycle Pu, compared to a once through scenario.

Fuel cycle back-end operations are a long-term issue and flexibility of the proposed scenario is an important feature to evaluate. To that end, its capability to deploy new technologies such as minor actinide recycle, the introduction of Fast Reactors, or future US waste disposal methods is assessed.

METHOD

Calculation tools

Available data on US UNF inventory was used to the maximum extent possible to produce a scenario as realistic as possible. Information on initial enrichment, burn-up, and cooling time was used to calculate radioactive inventory, including Pu isotopic composition. All corresponding calculations were based on Cesar, a reference code for used fuel recycling calculations.

To accelerate calculations, the following approximation was used. An Excel interpolation tool was developed and used; based on Cesar results, it was benchmarked and validated for PWRs with burn-up between 30 and 60 GWd/MTHM and initial enrichment between 3.7 and 5%. This validated domain covers most of the existing inventory, and uncertainties introduced by this approximation are deemed acceptable in the frame of this study. More complex calculations of MOX fuel evolution were done directly with Cesar.

The reference scenario (1) is the once-through cycle. In this case, waste inventory was calculated using only radioactive decay considerations.

MOX multi-recycling principle

Use of MOX fuels must provide the service (burn-up) expected by the reactor operator, with the required level of safety. To do so, the fissile material concentration (Pu-239, Pu-241) in the MOX must be high enough to maintain criticality in the reactor. For MOX fuel fabrication the current Pu limit for LWR fuels has an upper mass limit of 12.5% (safety constraint). Pu fissile isotopes are depleted in the reactor core, making it difficult to reuse directly the products produced by the recycling of MOX fuel. To compensate for Pu-239 depletion in used MOX, the large inventory of US UOx UNF is leveraged: one MOX UNF is blended with about 15 UOx UNF assemblies.

A second constraint was to keep the Pu-238 content in recycled MOX low enough to prevent excessive heat load and neutron emission during recycle operations. This is an issue for Pu multi-recycling since Pu-238 tends to accumulate in MOX. Current constraints in the MELOX MOX production facility impose keeping Pu-238 below 3.4% of total Pu. This limit is related to the specific design standard for the MELOX plant and could be reviewed / changed for a future plant. For the purpose of this study the current plant constraints were arbitrarily applied, to avoid making any assumption beyond the demonstrated reach of current technologies.

Pu-238 is initially produced in UOx fuels, mostly from U-235 via Np-237 (“the U-235 Way” as noted in Figs. 1 and 2) , but also from U-238 through Pu-241 beta decay to Am-241, neutron capture to Am-242, beta decay to Cm-242 decay, and alpha decay to Pu-238 (“the U-238 Way” as noted in Figs. 1 and 2).

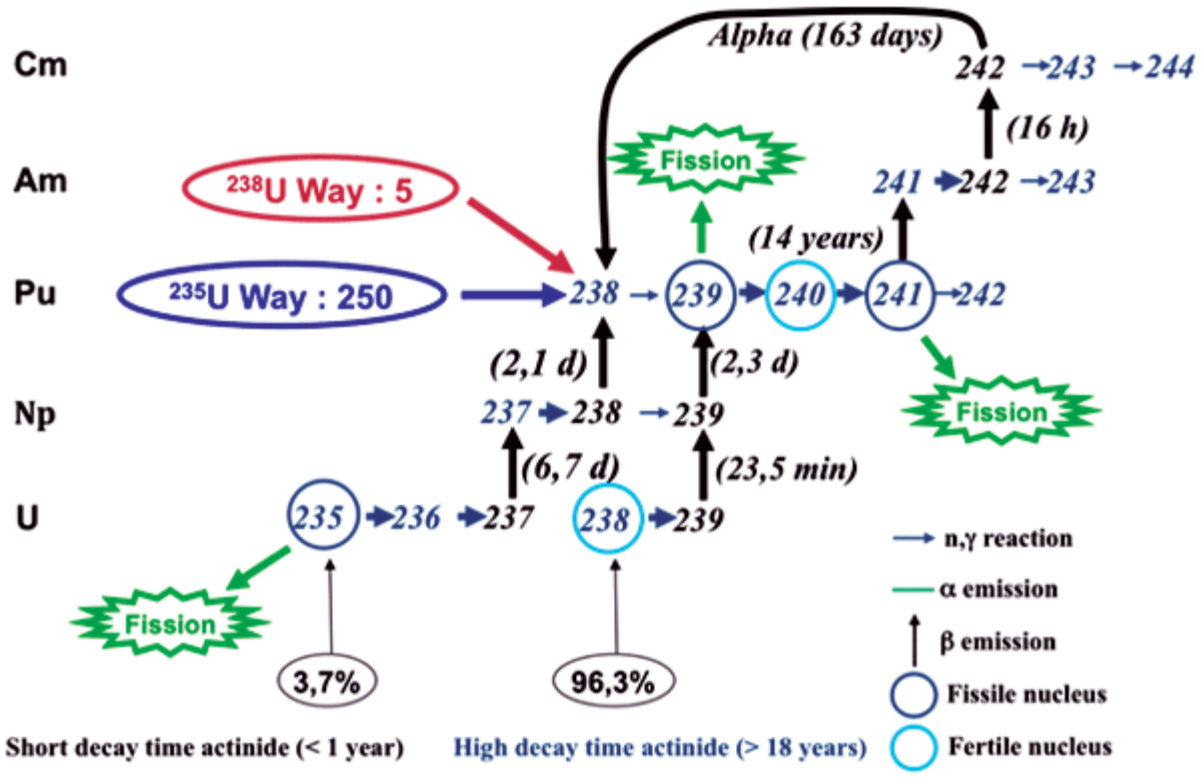

Figure 1. Pu-238 production in LEU (45 GWd/t)

In a typical MOX fuel, Pu-238 is generated directly from Pu-239, Pu-240, and mostly from Pu-241 introduced in the reactor ("the Pu-241 Way" as noted in Fig. 2). The initial Pu-238 isotopic concentration in the plutonium is about 0.2%. After a typical 45GWd/t irradiation and associated cooling time, about 0.173% is left in the used fuel, but 0.043% more has been generated from Pu-241/Am-241, leading to a net Pu-238 increase.

However, a MOX produced from aged UNF will contain a lower amount of Pu-241, thereby reducing the production mechanism of Pu-238. This leads to a net decrease of Pu-238 as indicated in Fig. 2. This shows that letting LEUOx UNF cool for an extended period of time helps relieve the Pu-238 constraint.

The longer cooling time does cause an increased amount of Am-241 in the HLW (from Pu-241 decay) and increases the benefit of minor actinide recycling in future advanced reprocessing. Also, in a future fuel fabrication plant, a Pu-238 constraint can be relieved through improved plant design.

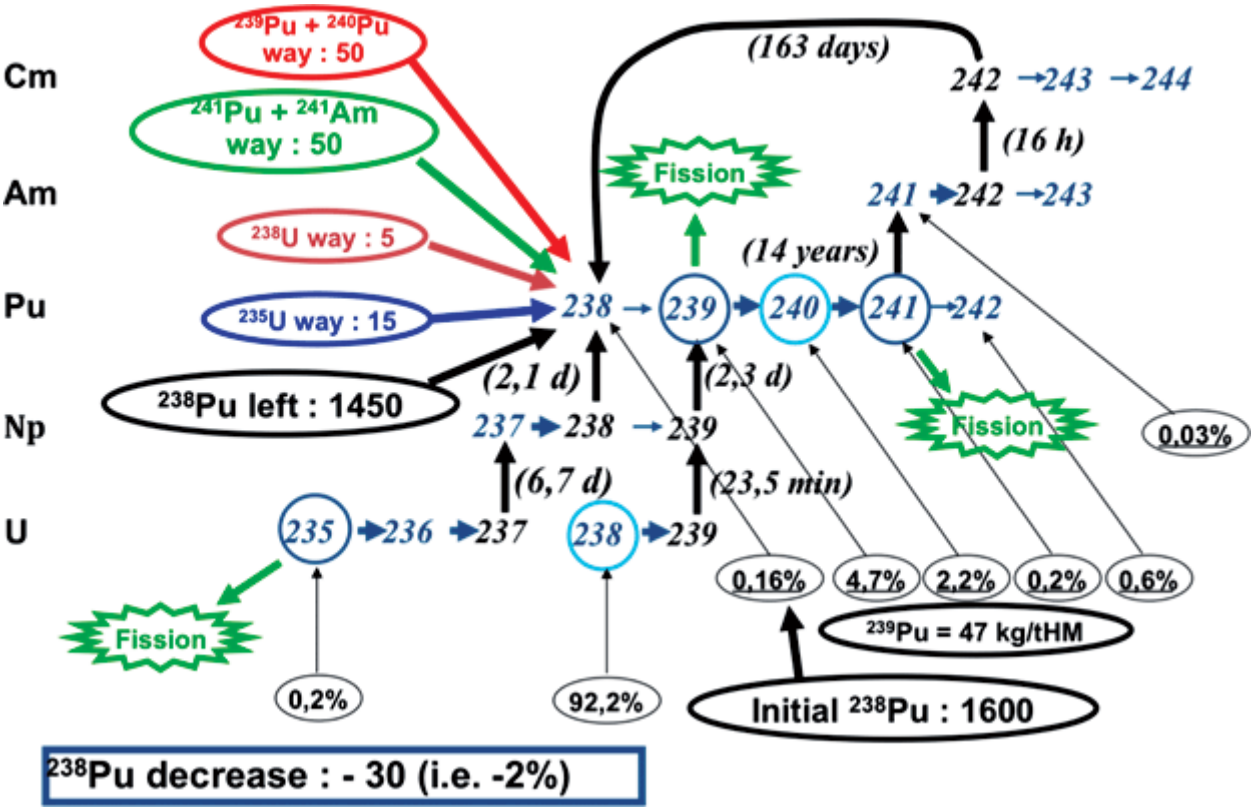

Figure 2. Pu-238 production in 45 GWd/t MOX fuel – 27yrs UOx cooling time

Recycling scenario timeline

The recycling scenario was built carefully. A first pilot plant is assumed starting in 2030. With a capacity of 800 MTHM/yr. the recycling plant would fabricate approximately 80 MT/yr of MOX fuel. MOX is recycled into the existing LWR fleet.

For the purpose of this study, the reactors where MOX is recycled are assumed to be EPR^{TM}'s. This was assumed in order to simplify the scenarios. Because of its improved design, the EPR^{TM} reactor presents the unique feature of being able to accommodate a 100% MOX core, with a constant Pu concentration over the fuel rods, thus simplifying MOX fabrication from what would be required for partial cores in an LWR.

For the purpose of this study, it was calculated that each EPR^{TM} is able to use 30MT/yr of MOX fuel, with an initial loading of 120MT of fuels. From a utility perspective, the use of such EPR^{TM}'s limits the number of reactors to be licensed for MOX. A first EPR^{TM} is loaded when 120 MT of MOX have been produced, and subsequent EPR^{TM}'s are loaded with MOX when MOX fuel is available. Over the course of the scenario, up to 6 EPR^{TM}'s are loaded with 100% MOX.

About ten years elapse between uranium extraction and the movement of used fuel from cooling pools at reactor site. In a similar way, about thirteen years elapse between UNF treatment and the corresponding recycled nuclear material again being available for treatment after one more cycle in a nuclear reactor, including 4 years in a reactor's core and 7 years cooling time (Figure 3). A 13-year period has been assumed in this study between the time a fresh MOX fuel is introduced in a reactor and the time recycled material from this same used

MOX is re-introduced. Another approach, with a longer MOX cooling time, has been studied in [2], leading to a longer approach to equilibrium and a larger inventory of cooling MOX fuels, but with less minor actinides in them.

In MOX UNF, the fissile plutonium isotopic concentration is depleted. To produce new MOX fuels, blending of the plutonium from MOX UNF with plutonium from LEUOx UNF, approximately 15 LEUOx UNF assemblies per MOX UNF assembly, is required to maintain sufficient fissile plutonium concentration. Also, treatment capacity has to be increased to provide adequate blending. MOX fuel production is also increased to accommodate the increased Pu stream. A 1,600MT/yr treatment capacity is assumed, doubling the original capacity, and another capacity increase to 2400 MT/y is eventually added. This ultimately enables processing all UNF that is being generated. It has to be noted that this timing is based on an "as soon as possible" assumption. A delayed availability of the higher capacity recycling plant would simply delay recycling used MOX fuels a few years. A two-step increase has been accounted for, with a conservative five-year ramp-up.

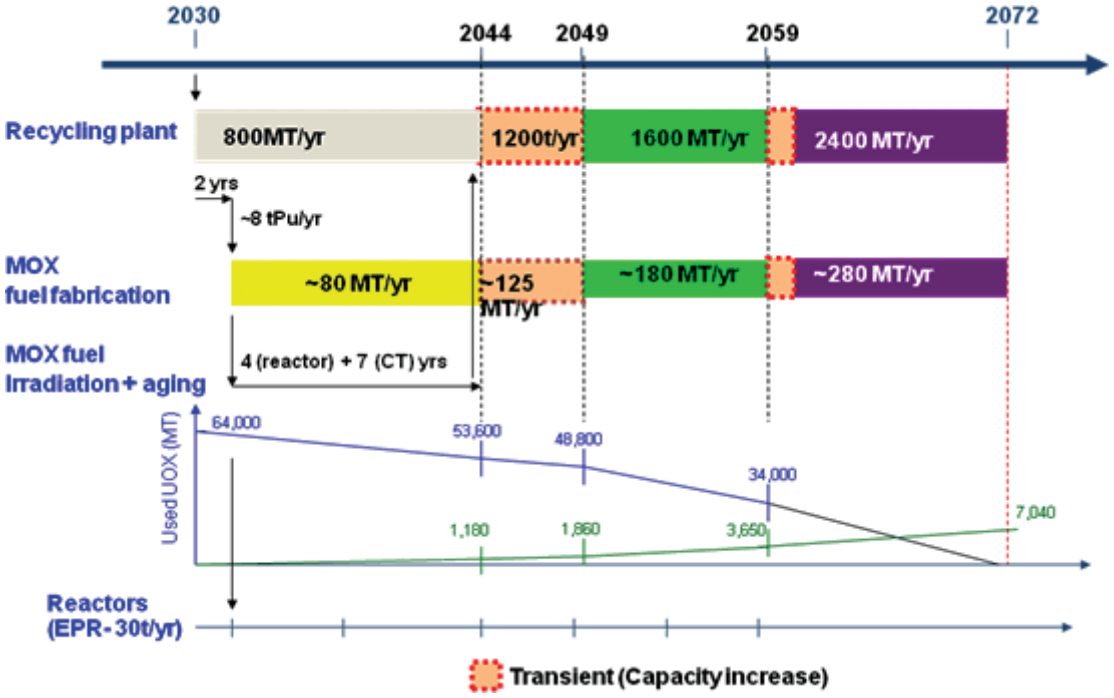

Figure 3. Timeline of the recycling scenario

For the purpose of this study an artificial constraint was imposed for case 2a and 2b that stopped the calculations once the whole initial inventory of the LEUOx UNF had been treated (64,000 MT in 2010 --- 110,000 MT in 2030).

At this point more than 7,000MT of MOX would have been fabricated and produced energy. Of this amount, 2,800 MT of MOX would have been already recycled in LWRs.

Waste inventory calculation and comparison with once-through cycle

At the end of the recycling scenario, after the amount of the 2010 U.S. inventory of 64,000 MT UNF had been treated, the waste inventory is assumed to be composed of canisters of vitrified HLW and compacted hulls/end pieces produced during treatment of the UNF. In addition, 4,200 MT of MOX UNF would be either still in reactor cores or during their 7 year cooling time and waiting for treatment.

Two cases have been considered. The first case (scenario 2a) assumes recycling will continue indefinitely, using GEN(IV) reactors assuming they are available in capacity to burn all plutonium from MOX UNF (about 24 MT/year of Pu). Another variant would be to divert to FRs only the excess plutonium (about 12MT/yr), to limit the necessary blending and cap the required recycling capacity. If GEN(IV) reactors are not yet available at that time, it is also possible to use U-235 to compensate for the decrease in fissile plutonium, with a constant treatment capacity [2]. Waste inventory is calculated assuming the used MOX fuels are treated, glass canisters and compacted canisters are produced, and Pu is recycled in available reactors. In scenario (2a), waste contains all UNF components except uranium and plutonium. The second case (scenario 2b) assumes a decision is made to stop recycling at the end of the scenario and send the residual UNF to a geological repository. In this case, the MOX UNF and LEUOx UNF assemblies are considered waste. This is done for comparison purposes, to determine the impact of a decision to stop recycling.

The waste generation, heat load, and radioactivity of the wastes from the once-through scenario (case1) and the recycling scenarios (2a) and (2b) were compared in this study. For the once-through scenario, the 64,000 MT of the initial inventory in 2010 were considered waste. In addition, to produce the same energy as produced by recycled materials in (2a) and (2b), additional UOx fuels would have had to be burnt at 45GWd/t. Waste comparison is made for the same level of produced energy. In the case of the recycling scenario, it is the energy produced by the initial inventory, plus the energy produced from the 7,000 MT of MOX. In the case of the once-through scenario, it is the energy produced by the initial inventory, plus additional 7,000 MT of 45GWd/t UOx fuels.

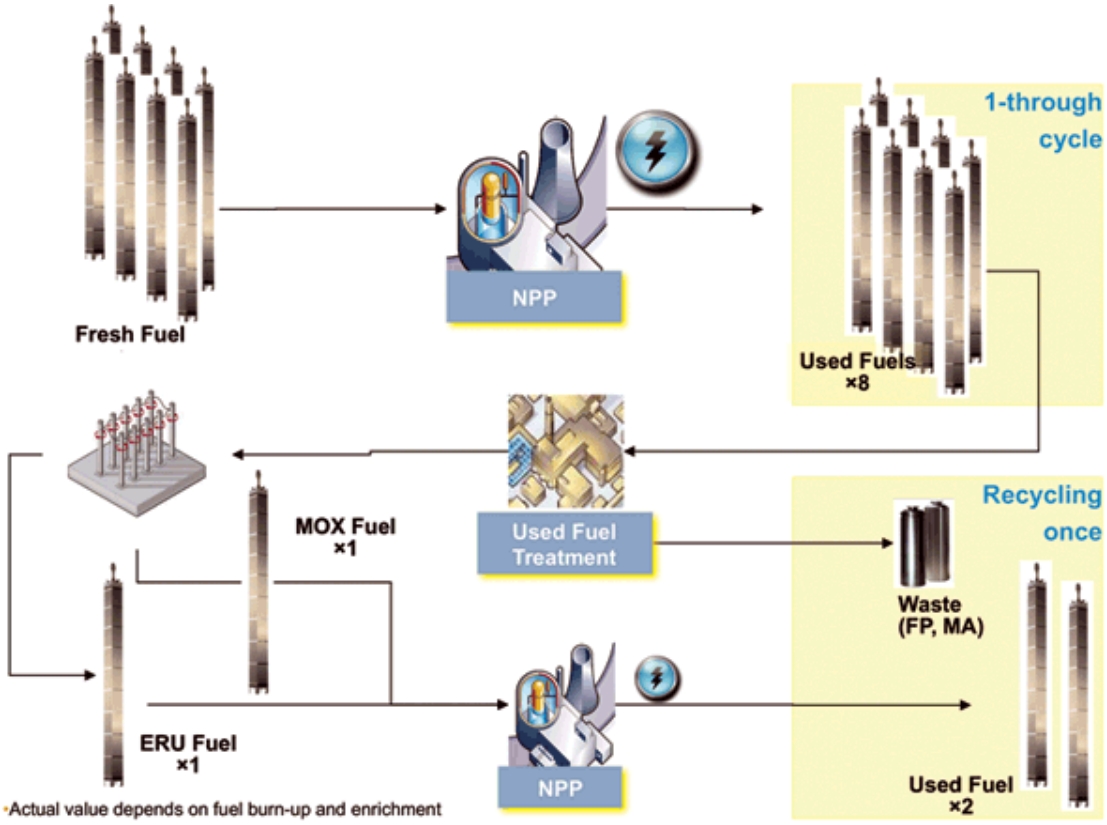

Figure 4. Waste inventory – comparison between once-through cycle and recycling

Advanced separation is not assumed for the duration of the recycling scenario. Major impact of Am separation on waste heat load and toxicity is therefore not credited in the recycling scenario, although it is very reasonable to believe such technology would be available by the time the first MOX fuels are treated (>2040).

RESULTS

Scenario case 1 (direct disposal) and recycling scenario cases (2a) (continuous recycle) and (2b) (limited recycle, then direct disposal) are compared in the following three figures , showing Pu mass, heat load and total alpha activity of the waste.

It was found that scenario (1) and scenario (2b) are fairly comparable from a waste load perspective. Scenario (2a) exhibits significant differences, in the mass of Pu immediately, and on heat load and alpha activity after about 1,000 years.

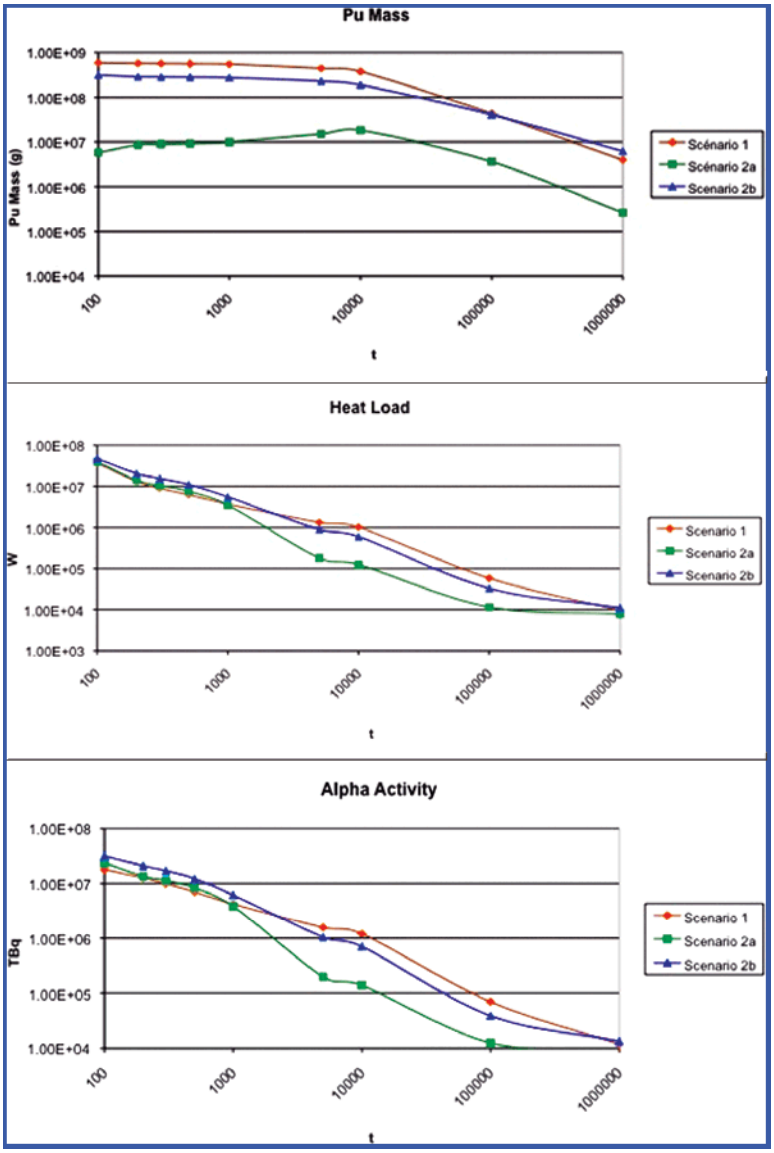

Figure 5. Pu mass, heat load and alpha activity in the waste, scenario (1), (2a), and (2b)

In the recycling scenarios, 64,400 MT of LEUOx UNF will have been treated, leading to the production of 7,040 MT of MOX fuels. And 2,800 MT of MOX UNF will have been recycled (40% of the production), the rest being in the reactor core producing energy or in the pools (cooling time) at the end of the scenario. Vitrified HLW canisters and compacted metallic waste canisters would be stored, but with a much reduced volume and no fuel integrity or security issues in comparison to direct disposal of the LEUOx UNF inventory.

The treatment capacities assumed in the recycling scenarios are not high enough to stop growth of the overall inventory of UNF in the U.S., except during the last 10 yrs. However, the net effect on UNF inventory is to have replaced 64,400 MT of existing aged used fuels, plus 7,040 MT of additional used UOx fuels that would be required without recycling, by 4,200 MT of MOX fuels less than 10 years old. In the non-recycling scenario, 64,400 MT of aged fuels would have to be stored, plus 7,040 MT of more recent UOx fuels, among which 4,200 MT would also

be less than 10 year old. During the same time (2030 – 2072 in the scenario considered), 84,000 fuels will have been discharged from reactors.

DISCUSSION

Results show the flexibility of the recycling scenarios. For the duration of these scenarios, Pu is managed in a way to avoid accumulating MOX UNF in the reactors pools, with or without GEN(IV) reactors. At the end of the scenario case 2b, a decision is assumed to be made to implement direct disposal as the preferred UNF management, results of this analysis show that the recycling scenario is fairly similar to the geologic repository waste load (total heat, total alpha activity, Pu mass).

During the time frame of the scenario case 2b, UNF accumulation will be reduced. More importantly, options to continue recycling will be kept open and minimize long term fuel mechanical integrity issues that otherwise could force direct disposal of part of the aging inventory. It will also have saved mining and enrichment of more than 59,000 MT of natural uranium, and will have reduced the volume of HLW. The need to open a geologic repository would be delayed, with secure waste canisters safely cooling in interim onsite storage.

Additional benefits are permitted by implementation of advanced technologies. To begin with, introduction of FR's before the end of the scenario would continue and increase a significant reduction of mining and enrichment of natural uranium, and increase utilization of the tremendous energy contained in depleted uranium. In a world where uranium is still accessible at a reasonable price, FRs may not be extensively deployed, except in countries with the political will to access full energy independence.

In the recycling scenarios, transition from LWRs to FRs can be accommodated easily, with recycled depleted and reprocessed uranium and plutonium being used to fuel the FRs which will operate with a reduced generation of minor actinides, in comparison to recycle in LWRs . It should be noted that, without Pu recycling in LWRs, the stream of Pu to be absorbed by the FRs would be doubled, requiring a doubled installed FR power capacity. Thus, the combination of recycle in LWRs and FRs is necessary for an efficient transition period.

Recycle of minor actinides will reduce the long term HLW heat load and radiotoxicity especially those generated in LWRs from irradiation of LEUOx and MOX fuels. When technology is available, introduction of minor actinide recycle, in addition to uranium and plutonium recycling, and together with the benefits of radioactive fission product decay before and after irradiation will have major impacts on capacity and residual hazards of the residual wastes emplaced in geologic repositories.

By definition, this opportunity is available only with a near term decision to begin recycling UNF in the U.S. It is reasonable to believe that effective technology will be available when the first MOX fuels are to be treated (>2040), if not earlier when the 800 MT/year pilot plant is built. If necessary and as proposed earlier by AREVA, a processing module could then be added to the first pilot plant and subsequent treatment units to provide the additional capability. AREVA has already implemented a significant overhaul of the liquid and solid waste treatment capabilities at the La Hague site, a few years after UP3 active commissioning, to eliminate the need for production of bitumen [3] and grouted metal waste. In the case of deploying advanced separation capabilities, it would be a matter of preserving the routes to upstream and downstream processes.

In the case of transition from LWRs to FRs, the industrial supply chain is progressively deployed. This scenario fits also with a progressive commissioning of FR's, with a long transition period expected. In addition, depending on the economics of FRs, the final situation may never be a transition to 100% FR fleets, but a combination of FRs and advanced LWRs. In this situation, the recycling scenario presented here would not only be a transition to a new era, but the first module of a long lasting symbiotic scenario, with Pu from LWRs being used in FRs, and Pu from FRs used in LWRs.

The present study casts some interesting light on a transition period between now and the beginning of GEN(IV) reactor deployment, and shows the benefits of recycling, whether the final decision is to move to GEN(IV) technology or not. Additional studies are still needed to build a transition scenario that includes minor actinide management.

REFERENCES

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[2] Emory D. Collins, Guillermo D. Del Cul, John P. Renier, and Barry B. Spencer, "Preliminary Multicycle Transuranic Actinide Partitioning-Transmutation Studies", ORNL/TM-2007/24, Oak Ridge National Laboratory (February 2007).

[3] Illustration of significant La Hague modification