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Americium mono-recycling in PWR: A step towards transmutation

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ABSTRACT

In contrast to the straight final disposal solution, countries like France have opted to reprocess their nuclear reactors spent fuel and defined another way to take care of sensitive elements such as the plutonium or minor actinides. Even in countries which have chosen to reprocess their spent fuel, americium is still considered as a final disposal waste. Among the minor actinides, americium will remain the main contributor to the toxicity and the decay heat of the spent fuel for thousand of years. Therefore it is important to reduce its quantity. At this time, only fast neutron future reactors are accepted to be efficient enough to transmute the americium from the thermal reactors spent fuel. As we can presume these future reactors will not be available before many decades, a new strategy which consists in recycling americium together with plutonium in pressurize water reactors mixed oxide fuel is proposed. In this paper the benefit and after-effect of this waiting strategy is analyzed. It demonstrates that the americium is indeed transmuted in a PWR quite efficiently (transmutation rate of around 43%) however the spent fuel is, as expected, more concentrated in curium of heavier nuclei. The impact on the fuel cycle (transportation, cooling time) is investigated showing that the key point would be the fabrication of the MOx-Am fuel.

1. Introduction

Minor actinides transmutation (especially americium transmutation) is usually devoted to generation IV reactors or Accelerator Driven Systems, pushing into the future this possibility. Though, transmutation only goes with multi-recycling and consequently the accumulation of heavier elements (Bk, Cf) which challenges the fuel re-fabrication (Griffin-Chahid et al., 2006). So, the fuel cycle needs an upgrade as well. Sodium cooled fast reactors (SFR) are supposed to be able to handle fuel loaded with americium either in dedicated blanket, either diluted homogeneously in the fuel (OECD, 2012; Taylor, 2015). Scenarios that consider this option reckon that the possibility to transmute americium into SFR will only be chosen when this technology will be available, implying that all americium produced in current light water reactors (LWR) would be considered as ultimate waste (DEN, 2012).

Regarding the French nuclear fuel cycle, the plutonium from spent UOx fuel, irradiated in Pressurized Water Reactors (PWR), is recovered via the PUREX (Plutonium and Uranium Recovery by EXtraction) process in order to build the MOx fuel, whereas americium is dissolved into the glasses with the fission products for the long term storage. Spent MOx fuel are then stored temporally as a source of plutonium that may be used into future SFR when those will be deployed.

Typically, one electrical gigawatt PWR produces about 3.1 kg of americium per electrical terawatt hour produced (TWhe). This number takes into account the reuse of the plutonium into the MOx fuel supposing that one MOx assembly needs the plutonium from height spent UOx. Americium net productions for different reactors are summed up in Table 1 (Sala, 1995).

Considering the waste produced by the French fleet and without taking into account the americium contained in the spent MOx fuel leaves approximately 40 tons of americium in the French glasses around 2020 (DEN, 2012). In a typical 100 GWd/t SFR, the americium production is around 3.9 kg/TWhe without any transmutation strategy (Brizi, 2010), meaning that the cumulative waste of 80 years of PWR operation in France (with 400 TWhe produced each year) would represent approximately 40 years of SFR

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Table 1
Americium production in different types of reactor (Sala, 1995).

Total Prod (kg/TWhe)	UOx (33 GWd/t)	UOx N4 (47.5 GWd/t)	MOx (43.5 GWd/t)	SFR (50 GWd/t)
^{241}Pu	4.6	3.6	24.6	7.6
^{241}Am	0.8	1.1	8.7	4.3
Total Am	1.2	1.7	14.2	4.7

operations (with the same electricity production). Accordingly, one has to wait for a long time before seeing any quantitative impact on the cumulated americium produced as waste if americium transmutation is chosen for future SFR. This fact has been demonstrated in much more details in Coquelet et al. (2009) which shows that the benefit of the transmutation would be far greater if we consider a possible future transmutation for the americium that is already produced in PWR. Assuming that a temporally storage of pure americium is not likely due to different safety reasons, and assuming that waste vitrification is an irreversible process, this work explores an alternative strategy to the MOx, by mono-recycling americium with plutonium, that would allow a maximum flexibility for the future fuel cycle in terms of radiotoxicity reduction. The goal is to keep the americium mass flow inside the fuel cycle for a possible incineration when capable technologies will be available.

Moreover, removing americium from the glasses will also impact the storage facility as the americium will considerably dominate the radiotoxicity of the waste accumulated in 2020 as shown in Fig. 1. This figure also represents the cumulated waste where americium would not be considered as a waste.

Today, the partitioning of minor actinides from the spent fuel (with a transmutation goal) is being addressed by mainly two processes, both as an upgrade of PUREX and relying on extensive R&D work. There is the so-called Grouped ActiNides EXtraction (GANEX) which will allow a grouped actinide separation by a selective extraction of Uranium and then partitioning of actinides from the fission products and lanthanides (Miguirditchian et al., 2008; Bell et al., 2012), this method is especially suitable for homogeneous recycling. Recently, a focus was put in the feasibility of separating americium alone, a liquid–liquid extraction EXAm process was developed and allow a selective americium separation from PUREX raffinate (Bollesteros et al., 2012). While those two processes have been demonstrated and technical feasibility can be considered as achieved, there is still some challenges (origin of selectivity, solvent cleanup,...) to overcome before industrial application.

In the scope of this work, as the idea of an americium separation without any direct use (dedicated systems for transmutation are not ready yet) of this material seems unreasonable, we can imagine using americium with plutonium for an innovative MOx-Am fuel in substitution of the actual MOx fuel. For example, the GANEX process could be upgraded to allow a U-Pu-Am phase, the americium from EXAm could also be mixed with plutonium. This paper focuses on the impact of using a MOx-Am fuel in present nuclear reactors. Consequences on a full PWR fleet, simulated with the code CLASS (Core Library for Advanced Scenario Simulation) (Core Library for Advanced Scenario Simulation, 2015), are also studied. It shows that the global americium production does not change a lot, despite the high americium transmutation rate observed in the PWR.

2. Depletion calculations and assembly model

In order to study this alternative strategy based on MOx-Am fuel to the current MOx fuel where americium is directly put into the waste glasses, the reactor physics of both fuel possibilities as well as the effect on the fuel cycle is studied. CLASS takes advantage of meta models, therefore the philosophy of this study will be identical to the one followed in Leniau et al. (2015b) which is based on numerous single assembly evolution calculations (described below) that are then used to build a numerical model in order to predict the fissile material proportion which allows to reach a given Burn-up. Those different calculations are also used to build an evolution predictor for the inventories of the nuclei under irradiation, the goal being to predict, for any burn-up and any fresh fuel composition, the isotopic composition of the fuel during its irradiation. So, whatever the burn-up of the fuel, we know how its composition will evolve with time.

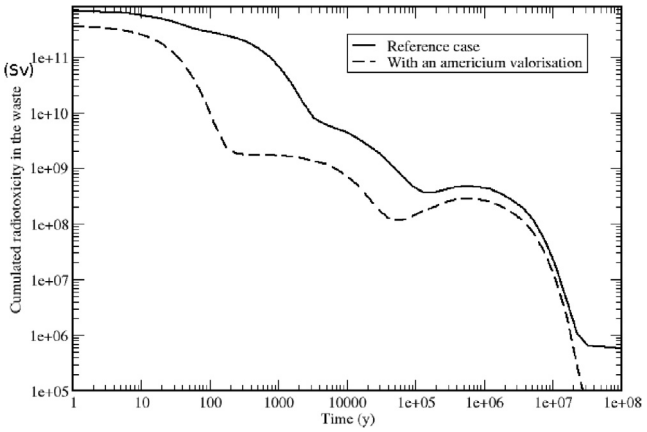

Fig. 1. Radiotoxicity of the accumulated waste in 2020 for the French fleet regarding different strategies: current strategy (solid lines) where the americium is sent to the waste and an alternative strategy where americium is not located in the glasses (dashed line).

2.1. The assembly model

Usually, the study of a reactor irradiation is performed in two steps: a cell calculation and a core calculation. Here, we assume that the MOx/MOx-Am composition will not affect core aspects of the reactor physics (like neutron leakage or reactivity follow-up with boron for instance). We then focus on the cell or assembly level. This is an approximation which bears significant biases that need to be addressed very carefully (Somaini et al., 2016; Kepisty et al., 2016), however it is found to be a good compromise between desirable precision and computational cost. Each cell calculations have been performed with the MURE package (Meplan et al., 2009), available at the NEA software databank. MURE is a depletion code that calculates the evolution of any isotope under neutron irradiation. The neutron properties (neutron reaction rates with nuclei) are calculated at different time with the use of MCNP (Briesmeister, 2000). The integration of the evolution equation (so-called Bateman equations) is performed thanks to a *Runge & Kutta 4* method. The use of a stochastic neutron transport code allows the simulation of various fuel compositions without any concern of the neutron self-shielding modeling issues. The simulated geometry is a typical PWR assembly in asymptotic irradiation conditions (mirror boundaries surrounding the geometry).