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Neutronic and fuel cycle comparison of uranium and thorium as matrix for minor actinides bearing-blankets

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ABSTRACT

Minor actinides transmutation is one of the three main axes defined by the 2006 French law for nuclear waste management, along with long-term storage and use of a deep geological repository. In the heterogeneous approach, minor actinides are loaded in specially designed targets assemblies which are located in the periphery of the core, in order to limit the impacts on core operations. In this paper, we compare the use of uranium and thorium dioxide as support matrix in which minor actinides are diluted in the target assemblies. Both UO_2 and ThO_2 exhibit sufficiently good irradiation behavior to withstand the long residence time associated with heterogeneous transmutation. Five different reprocessing strategies are compared in which some or all the elements in the blankets are reused after reprocessing. The impacts on core safety parameters and fuel cycle parameters are also evaluated for each case and it is found that using thorium as support matrix with reuse of uranium 233 leads to transmutation performances similar to the one obtained with the reuse of plutonium from uranium blankets with slightly lower global impacts on reactor operation and fuel cycle.

1. Introduction

Minor actinides are a set of three main elements (neptunium, americium and curium) which are by-products of uranium irradiation in nuclear reactors. These elements are produced in relatively limited quantities (NEA, 1999) but they exhibit long-term radiotoxicity and decay heat levels which complicate the handling of associated nuclear waste.

In the case of a closed nuclear fuel cycle strategies where spent fuel is reprocessed and plutonium reused in fast spectrum reactors, minor actinides are the main contributors to long term radiotoxicity of the spent fuel and to decay heat of the ultimate waste package. Minor actinides transmutation has thus been proposed as a potential solution to decrease the burden of nuclear waste and to reduce the constraint on the final repository (Salvatores et al., 1995).

Transmutation in critical reactors can be done in two different ways:

- Homogeneous transmutation, in which minor actinides are directly mixed with the reactor fuel. This solution exhibits the best performances as the minor actinides are exposed to a high flux level. However, it exhibits the drawback of contaminating the entire fuel cycle with minor actinides and it decreases the "safety" performances of the reactor. Minor actinides content of up to 5%vol can be loaded depending on the considered core design. Additionally, the residence time of the minor actinides bearing fuels cannot exceed the one of standard MOX fuel.
- Heterogeneous transmutation in which minor actinides can be loaded in specifically designed assemblies, usually in the periphery of the core, which are called "Minor Actinides Bearing Blankets" or MABB. The use of such subassemblies helps decoupling minor actinides management from the fuel and thus gives a larger flexibility compared to the homogeneous mode. As these blankets benefit from the neutron leakage from the active zone they have almost no impact on the core neutronic parameters such as delayed neutron fractions or sodium void worth. This allows to load a large minor actinide mass and to reduce the number of MABB to be manufactured. On the other hand, the obtained performances are lower than the previous one as the flux level seen by the assemblies is quite low. Minor actinides content between 10% and 40% are expected to be loaded in such cores. As fuel and MABB cycles are decoupled, higher transmutation rates can be expected at the cost of longer irradiation time.

The present paper focuses on heterogeneous transmutation strategies. A thorough analysis of this transmutation approach

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has been carried out by a NEA task force in 2009 and summarized in NEA (2012). The main points are described below:

- The high content of minor actinides in the fuel requires important fuel design effort, notably in terms of mechanical design. Previous experiments, especially the SUPERFACT experiment in which pins with up to 45% of americium and neptunium were irradiated in the Phenix reactor core (Prunier et al., 1993) showed that MABB irradiation was accompanied by an important production of Helium due to alpha decay of minor actinides nuclei which has an impact on the mechanical behavior of the pin and on the size of the gas plenum.
- Power production in the MABB assemblies is also very low at the beginning of irradiation which puts tighter constraints on the mechanical design as fuel restructuration does not happen at low temperatures. The important power variation during irradiation also increases the strain on the fuel pins, possibly leading to thermal cracking. (Bonnerot et al., 2010)
- Decay heat, gamma and neutron emission of irradiated and refabricated MABB assemblies is significantly higher than for a standard MOX fuel, which leads to additional issues in terms of fuel handling, reprocessing and manufacturing.

When considering minor actinides transmutation, several objectives are usually pursued. Firstly, the transmutation performances, e.g. the amount of minor actinides which are effectively turned into fission products during irradiation is considered, as it is a direct estimator of the performances of the process. The support ratio, which is the number of reactors which production can be absorbed in one minor actinide burner, is also of interest from an economic point of view. The support ratio can be reduced either by decreasing the production of minor actinides in the reactor or by increasing transmutation performances, as discussed here.

It has been proposed to add moderating material in the MABB in order to increase the transmutation performances of the design (De Saint Jean, 1998). Slowing down the neutrons in the blankets has the interest of increasing the absorption cross sections and thus the number of captures or fissions. However, it also increases the amount of curium produced, which is more troublesome than americium on fuel back-end due to a higher neutron source and specific decay heat. This addition is especially interesting in case of once-through transmutation, in which the blankets are irradiated only once and then discarded as waste. We considered here a heterogeneous transmutation scheme in which irradiated blankets are reprocessed to maximize amount of transmuted material.

Similarly to plutonium, minor actinides cannot be loaded *per se* as oxide fuel in a reactor but must be blended with a matrix to produce usable reactor fuel. Several materials have been proposed as potential matrices for MABB fuels. The first one is evidently uranium dioxide (UO_2), which has been tested in the SUPERFACT experiments for instance. UO_2 is a well-known material with a low swelling rate and which can withstand the long residence time associated with heterogeneous transmutation. However, the use of uranium oxide as support matrix comes with a production of plutonium which may cause an issue in terms of proliferation. It also implies a modification of the core to keep a total breeding gain close to unity. An analysis of the impact (or lack thereof) of the use of minor actinides blankets has been done in Buiron et al. (2009). Additionally, the irradiation behavior of mixed uranium-minor actinides oxide fuel has yet to be fully characterized (IAEA, 2009).

Inert Fuel Matrix, or IMF, has also been discussed for transmutation in heterogeneous mode. A review can be found in Matzke et al. (1999). In this concept, the minor actinides are embedded as oxide in either a ceramic material (Cercer concept) or a metallic material (Cermet). This removes the production of plutonium due to capture by uranium 238 in the target but the selection of the matrix is complicated as it should exhibit a good thermal conductivity, acceptable swelling under irradiation and good irradiation resistance behavior to neutrons, alpha and fission products. No matrix has been found featuring all these parameters. However, a possible hybrid matrix of $AmZrO_2$ dispersed in an $MgAl_2O_4$ matrix, which limits damage to $MgAl_2O_4$ by fission products irradiation while making good use of its otherwise good stability has been proposed in Chauvin et al. (1999). Issues regarding dissolution of the inert matrix must also be addressed (see for instance Ebert et al., 2015).

Thorium has been proposed as a potential nuclear fuel in the Th232/U233 fuel cycle, in which fissile uranium 233 is bred from thorium 232. This cycle can be closed in fast or thermal reactors, although it requires an initial stock of fissile material (U235 or Pu239) to start the breeding process. The potential benefits coming from the use of this cycle are listed in IAEA (2005). To name but a few, this option virtually removes minor actinides production and increases the reserve in fertile material by a factor three to four as thorium is more abundant than uranium while being intrinsically proliferation resistant due to high gamma production of daughter isotopes of U232. India is currently the leading country for thorium fuel cycle industrialization. Thorium dioxide (ThO_2) or thoria is also a relatively well-known material which performances under irradiation are better than those of UO_2. A detailed study of thorium properties as a nuclear fuel can be found in Hania and Klaassen (2012).

Thorium dioxide use has been already discussed as support matrix for heterogeneous transmutation in once-through scheme, for instance in Lombardi et al. (2008). In this case, advantage is taken of the low solubility of thoria in groundwater for long-term storage. Additionally, this option limits the production of plutonium in the blankets, thus decreasing the total radiotoxicity at disposal. Advantage has also been taken of the lack of plutonium to transmute plutonium and minor actinides without separation during reprocessing, in the case of it being not acceptable. The use of thorium axial blankets in such a case and the related neutronic impacts are discussed in You and Hong (2014).

We elaborate in this paper on the possibility of using thorium dioxide (ThO_2) as a support matrix for minor actinides bearing blankets in the case of a multi-reprocessing scheme in plutonium-fueled fast reactors. We compare the relative performances of uranium and thorium for this application in terms of reactor and fuel cycle impacts. The methodology and tools used are detailed in the first part and the effects of thorium and uranium matrixes on reactors parameters, fuel cycle and transmutation performances are then analyzed in the following sections.

Several cases can be envisioned for transmutation with a thorium support. We considered that thorium was used in combination with a conventional U/Pu fuel cycle. The following possibilities for thorium use which were investigated here are:

- Thorium could be used only as support matrix and the bred uranium 233 can be recovered after reprocessing and used for starting an independent thorium/uranium cycle. As this cycle requires an initial supply of uranium 233 for starting, this solution would allow a reduction of the total inventory of minor actinides during the switch to thorium while producing the necessary uranium 233. Similarly, the uranium production could be incorporated in the reactor core as fuel, thus replacing part of the plutonium and decreasing the minor actinides production. This option was not pursued here.
- Uranium 233 produced during irradiation could be reused as a neutron supplier directly in the blankets, in order not to mix plutonium and uranium 233 in the standard fuel cycle. In this approach, plutonium from the blankets is recovered to be used