Annals of Nuclear Energy 97 (2016) 198–203

Contents lists available at ScienceDirect

Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene

Evaluation of the Americium transmutation performance in high flux reactors

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ARTICLE INFO

Article history:
Received 3 January 2016
Received in revised form 10 June 2016
Accepted 28 June 2016
Available online 21 July 2016

Keywords:
Transmutation
Americium
ChainSolver 2.34
High flux reactor
SM3 reactor

ABSTRACT

The numerical transmutation of Americium heterogeneous loaded for one cycle in thermal high flux reactor category was realized. The transmutation calculations are performed based on ChainSolver 2.34 code. A comparison with the measurement and calculation results of the burn up of ^{241}Am irradiated in HFR at Petten was examined to evaluate the accuracy of current available numerical tool. To reach the Am destruction with a short irradiation time, a high flux SM3 reactor having a flux density of thermal neutrons higher than 10^{15} cm^{-2} s^{-1} was proposed. To obtain transmutation rate of 99.75%, the Am samples needed only 90 exposure days in full power for SM3 reactor. The effectiveness results suggested an effective use of SM3 as compared to Petten HFR. Finally, the results have been discussed in order to propose a new concept of high flux reactor destined for the destruction of actinide minors, in particular ^{241}Am.

1. Introduction

Americium is one of the minor actinides that contribute to a large part of the radiotoxicity of spent fuel from power nuclear reactors. Its most abundant isotope (about 70%), ^{241}Am, decays with a half- life of 430 years to ^{237}Np by emitting an alpha particle, which dominates the long term leakage risks of underground repositories. So, it should be partitioned from the spent fuel and transmuted by neutron irradiation in nuclear reactor into fissile Plutonium and Curium isotopes, in order to reduce both the inventory and the radio-toxicity of radioactive waste. Fig. 1 shows the expected reduction of heat load in a repository. The multiple Pu recycling and minor actinides disposal has limited benefits (factor <2). The multiple Pu and Am recycling has a more favorable impact (e.g. factor ~5–6 at 1000 years after disposal) (Physics and Safety of Transmutation Systems, 2006).

The Americium transmutation has been studied and tested in different types of reactors including thermal and fast neutron fluxes, in order to perform its feasibility and its effectiveness, before it can eventually be introduced on an industrial scale.

At present, the consequences of Am recycling on the core safety and the fuel cycle are investigated for several reactor categories. Its homogeneous recycling is not recommended due to the severe impact on core performance and the contamination of the whole fuel cycle with Pu and Cm isotopes. But, its heterogeneous recycling is investigated by the EFTTRA collaboration. Both multiple recycling of Am in a fast reactor and once through irradiation cycle in high thermal or epithermal neutron fluxes are considered (Kloosterman et al., 1997). More details can be found in EFTTRA work documents, see the reference (Babelot et al., 1997).

The present paper, deals with the simulation of pure ^{241}Am samples transmutation in research thermal high flux reactor HFR at Petten (The Netherlands) to evaluate the conversion rate of this isotope in high flux neutron environment during the irradiation, and deals with the comparison with the experimental results of EFTTRA T4 and EFTTRA T4bis tests (see references Babelot et al., 1998; Konings et al., 2000 for more details) to examine the accuracy of current available numerical tool. The transmutation calculations are performed based on ChainSolver 2.34 code. This program is intended for carrying out fast transmutation calculations of samples during irradiation in nuclear reactors. Their calculations are extremely tedious because of at each stage of an irradiation the specific neutron fluxes, reaction cross-sections, irradiation time and the initial isotopic composition should be counted separately. This code use the ORIP_XXI program complex data file information. All details are given in the reference (Romanov et al., 2005).

To reach the Am transmutation with a short irradiation time, a simulation irradiation of a similar Am samples was effected in research thermal high flux SM3 reactor (Dimitrovgrad, Russia) having irradiation channels with a flux density of thermal neutrons higher than 10^{15} cm^{-2} s^{-1}. The effectiveness of Am transmutation in both Petten HFR and SM3 reactor were evaluated. Finally, the

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http://dx.doi.org/10.1016/j.anucene.2016.06.036
0306-4549/

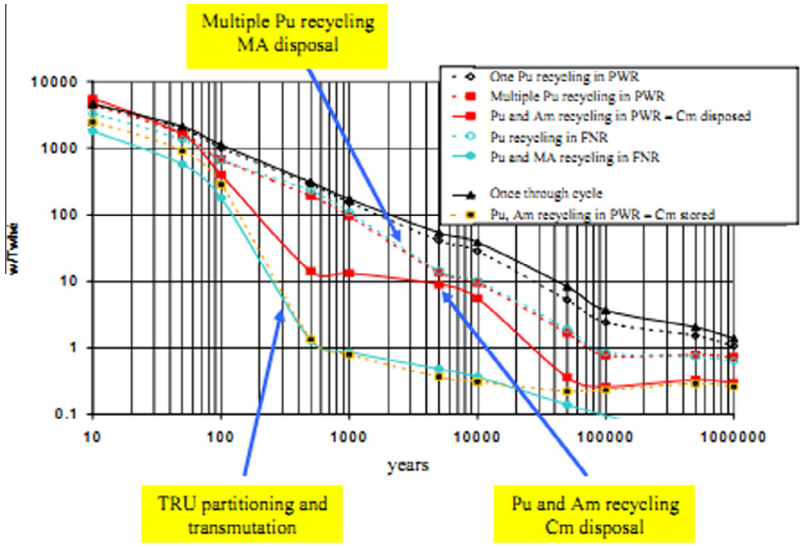

Fig. 1. Heat load in a repository.

results have been discussed in order to propose a novel concept of high flux reactor designed for the destruction of actinide minors, in particular ^{241}Am.

2. Am extraction from spent fuel

Americium can be extracted using the PUREX procedure in a complementary extraction process (Article Americium From Wikipedia, 2016; Afsar et al., 2014). After PUREX reprocessing of Uranium and Plutonium from spent fuel, the lanthanides and remaining actinides are then separated from the aqueous residue (raffinate) by a diamide-based extraction (Article Americium From Wikipedia, 2016). These elements present in solution in +3 valence state. It has been shown that many heterocyclic ligands containing soft N-donor atoms are capable of separating trivalent actinide ions (Am(III) and Cm(III)) from trivalent lanthanide ions Ln(III) Afsar et al., 2014; Suzuki et al., 2007. Am and Cm are difficult to separate because they both have the similar chemical proprieties. Their separation can be achieved by treating a slurry of their hydroxides, whereas Americium oxidizes to soluble Am(IV) complexes which can be separated from Cm(III) Article Americium From Wikipedia, 2016; Afsar et al., 2014; Suzuki et al., 2007. On the other hand, it possible to extract Am(III) from Cm(III) using chromatography employing organic resins (Afsar et al., 2014; Suzuki et al., 2007). Finally, metallic Americium is obtained by reduction from its compounds (Article Americium From Wikipedia, 2016).

3. Am transmutation in a thermal flux

The transmutation of long lived nuclides by neutron irradiation in nuclear reactors is the operation of their artificial transformation into stable or short lived elements. For radioactive minor actinides, there are two processes that they are successfully transmuted, one process is to absorb neutron and fission, another process is to capture a neutron and change to new radioactive transuranic nuclides (Liu et al., 2014).

The ^{241}Am isotope may be transmuted in the thermal reactors effectively by neutron capture reactions because it has very large capture cross section in a thermal neutron environment (see Fig. 2). Under irradiation, about 10% of it is converted to the fissile ^{242m}Am, while the remaining part is transmuted to ^{242}Am which decays with a half life of 16 h to ^{242}Cm by β^- emission (83%) and ^{242}Pu by electron capture (17%). The ^{242}Cm decays with a half life of 163 days to ^{238}Pu by α emission, and the ^{242}Pu creates ^{243}Pu by neutron capture which then decays to ^{243}Am. Through radiative capture, ^{238}Pu gives rise to the fissile ^{239}Pu. The ^{243}Am is not fissile and should be transmuted to ^{245}Cm by two successive neutron captures before it can be fissioned. For more details see references (Kloosterman et al., 1997; Babelot et al., 1997, 1998; Konings et al., 2000; Klaassen et al., 2002, 2003; Romanov et al., 2005; Article Americium From Wikipedia, 2016; Afsar et al., 2014; Suzuki et al., 2007; Lewis et al., 1309; Liu et al., 2014; Berthou et al., 2003) and Fig. 3.

In any case, several neutrons are needed to transmute the Am to fissile Plutonium and Curium isotopes. Consequently, the Am transmutation should perform in high flux reactor characterised by its high density of a thermal neutron flux. After adding ^{241}Am to the thermal HFR core, the thermal neutrons decrease and the high energy neutrons increase. So, it actually can act as burnable poison in the thermal reactors. It can be used to partially substitute for the burnable poison, reduce the concentration of the boric acid in the coolant and increase the negative temperature coefficient in the thermal reactors, all these measures can drastically reduce the possibility of the critical accident in a fuel cycle. The spatial self-shielding effects of the heterogeneous distributions of Am samples in the reactor core can avoid the initial reactivity to drop significantly after adding ^{241}Am, see the reference (Liu et al., 2014).

4. Am transmutation performance: A comparison of measurement and calculation results

In this study, thermal high flux reactor types are further designed to perform the Americium transmutation with high level burn up, considering the large capture cross section of ^{241}Am in thermal region. Both Petten HFR and SM3 HFR were proposed. The Petten HFR is a tank in pool type reactor. It operates at a constant power of 45 MW using low enriched uranium plate-type fuel, with Light water cooled and moderated. The reactor core has a total of 33 fuel rods, 6 control rods, 25 reflector rods and 17 experiment positions, see Fig. 4. The SM3 HFR is vessel type water cooled with a trap. It has UO_2 fuel with a high enrichment in U-235 90%. The thermal power makes up 100 MW. There are 30 channels 68 mm in diameter in the beryllium reflector, 27 channels