Possibilities for High Level Waste Reduction through Transmutation of Minor Actinides

K. Zashev

DTNPE, Technical University of Sofia, 8 Kliment Ohridski Blvd., 1000 Sofia, Bulgaria

Abstract. For many years there has been a sustained international interest in partitioning and transmutation of the actinides, and particularly americium, neptunium and curium, mainly because of the major contribution to the decay heat and radiotoxicity of spent nuclear fuel. Therefore in the current report are examined the possibilities for high level waste reduction through transmutation of these three minor actinides. The main objectives of minor actinide partitioning and transmutation processes are described and the technological options of such process are revealed. A software experiment showing the possibilities of actinides transmutation of the most utilized thermal reactors is conducted. The experimental results and comparison of the transmutation possibilities of thermal reactors is also shown.

Keywords: minor actinides, spent nuclear fuel, thermal reactors, transmutation.

1 Introduction

The word transmutation originates from the never-realized goal of ancient alchemists to transform, or transmute, the base metals into gold. Literally transmutation means: to change or alter in form, appearance, or nature; to change something completely, especially into something different and better.It was first consciously applied to modern physics by Frederick Soddy when he, along with Ernest Rutherford, discovered that radioactive thorium was converting itself in to radium in 1901. When it comes to nucleus, nuclear transmutation is the conversion of one chemical element or isotope into another. In other words, atoms of one element can be changed into atoms of another element by 'transmutation'. This occurs either through nuclear reactions (in which an outside particle reacts with a nucleus), or through radioactive decay (where no outside particle is needed).

For many years there has been a sustained international interest in partitioning and transmutation of the minor actinides neptunium, americium and curium produced by fission reactors. Although these three elements are produced in relatively small quantities in fission reactors, they are major contributors to the decay heat, neutron output and radiotoxicity of spent nuclear fuel [1]. Neptunium, americium and curium are intensely radioactive, featuring both alpha decays and high energy gamma emissions and dominate the total radioactivity of spent nuclear fuel orvery high level waste once the fission products have decayed at about 500 years after discharge from the reactor.

Transmutation of the minor actinides by fission or neutron captures to produce shorter lived products would reduce the burden of radioactivity in a geological repository. Many consider that this would make nuclear fission more acceptable to the public and this is the motivation for studying minor actinide transmutation [1]. A nuclear fuel cycle with minor actinides extracted from spent nuclear fuel and re-irradiated in a reactor could in principle lead to a reduction of up to a factor of 100 of the long term radiotoxic burden. Some countries, especially France, Germany and Japan, see minor actinide transmutation as a very important goal and have made very large research and development commitments to it [1].

2 Minor Actinides Transmutation Obectives

Partitioning and Transmutation of actinides are generally said to have three main objectives [2]:

1. *Reduction of the possible dose to the population due to leakage of actinides from underground disposal sites;*
2. *Reduction of the total radiotoxic inventory of underground disposal sites;*
3. *Using fissile actinides for energy production.*

Each of these three objectives has its specific impact for the partitioning and transmutation strategy. The first objective implies that the initial inventory of Np-237 and its precursors shall be reduced, because Np-237 is a nuclide which is easily transported to the biosphere without much adsorption in the underground once it is released from the underground disposal site. Studies on underground disposal options in salt domes show that Np-237 dominates the possible dose to the population living some millions of years from now.

The second objective is met by heavily reducing the total amount of actinides in High Level Waste (HLW). It is believed that reducing the radiotoxic inventory per disposal site or reducing the number of disposal sites has some positive impact on the public opinion with regard to the nuclear waste problem. It also leads to lower probabilities of human intrusion in repositories and to reduced accompanied doses received by men after intrusion. For human intrusion scenarios the contribution of americium to the dose is dominant. Also from the economic point of view it is advantageous to reduce the inventory or the number

of underground disposal sites to be licensed. This can be accomplished also by reduction of the heat emission of the waste.

The third objective is met by fissioning of actinides in nuclear energy parks with thermal and fast reactors. This was the main reason to apply reprocessing. Future incentives for partitioning of actinides depend primarily on future prospects of nuclear energy, which are highly affected by political standpoints.

3 Radiotoxicity of SNF

The harmful potential of a certain quantity of radioactive material is evaluated using the concept of *radiotoxic inventory* and the term of potential *radiotoxicity*. The radiotoxicity of a nuclide is determined by the productof the activity and the effective dose coefficient '**e**' for the given isotope. Effective dose coefficient corresponds to the doseresulting from the intake of 1 Bq of a specific radionuclide. The quantity T is the integration time in years following intake. For adults, the integration time is 50 years [3].

$$\text{Radiotoxicity} = \text{Activity} \cdot e(T)\,.$$

The activity A is the number of decays per unit time interval and it is measured in Becquerel, Bq [3]:

$$A = -\mathrm{d}N/\mathrm{d}t = \lambda N\,.$$

Effective dose coefficients are evaluated using models describing the movement of radionuclides through the different parts of the body. A dose factor table is regularly updated by the International Commission on Radiological Protection (ICRP).

$$\begin{aligned} &\text{The ingestion radiotoxicities}: \quad RT_{\text{ing}} = Ae_{\text{ing}}(50), \\ &\text{The inhalation radiotoxicities}: \quad RT_{\text{inh}} = Ae_{\text{inh}}(50)\,. \end{aligned}$$

Two basic approaches are used for creating a broader vision for radiotoxicity. The first approach is used by International Atomic Energy Agency and is related on Safety Guide RS-G-1.7 – 2004 [4] which use the concept of "clearance". The clearance is intended to indicate which material under regulatory control can be removed from this control. Clearance is defined as the removal of radioactive materials or radioactive objects within authorized practices from any further regulatory control by the regulatory body. Further, the clearance potential index is defined as ratio between specific radioactivity and clearance level, the state in which the radioactivity material became undangerous. So, the clearance potential index is an nondimensional quantity, which, if greater than one, shows the measure of radiotoxicity for the radioactive material and when is equal to one or less means that the radioactive material became clean in the sense that can be manipulated without restrictions.

The second approach, or the US approach, is described in the Code of Federal Regulations [5]. The code provides the radioactivity concentration guides (RCG) for continuous ingestion (from water) and inhalation (from air) in unrestricted areas, in units of curies per cubic meter (Ci/m^3). The RCG values specify the maximum permissible concentrations of an isotope in soluble and insoluble forms, for both ingestion and inhalation, and for occupational and unrestricted exposure. When the activity (in curies) of a given isotope is divided by the radioactivity concentration guides for that isotope, the result is the volume of water (or air) required to dilute that quantity of the isotope to its maximum permissible concentration. The dilution volume is a measure of the radioactive toxicity of the nuclide for cases of direct ingestion or inhalation and is known as ingestion or inhalation hazard factor.

4 Technological Possibilities for Minor Actinides Transmutation

There are several types of variants for minor actinides transmutation:

- **Acceleration-Driven Systems** – The transmutation of long-lived radioactive waste can be carried out in an accelerator-driven system (ADS), where neutrons produced by an accelerator are directed at a blanket assembly containing the waste along with fissionable fuel. Following neutron capture, the heavy isotopes in the blanket assembly subsequently fission, producing energy.
- **Fusion-fission Reactors** – Fusion–fission hybrid reactor is a new energy system based on a fusion reactor [6]. The fusion–fission hybrid was conceived to capitalize on the advantages and minimize the disadvantages of both processes (fusion and fission). Fusion reactors are 'neutron rich' and 'power poor' while fission reactors are 'neutron poor' and 'power rich'. The idea is to build a hybrid device, the core of which consists of a fusion reactor whose purpose is to supply a steady flux of neutrons to a surrounding blanket of fissile materials. Such a reactor could generate electricity, produce fuel for conventional fission reactors or provide a way to transmute the long-lived actinides of nuclear waste into shorter-lived and materials that are more safely disposable [7].
- **J-PARC** – Japan Proton Accelerator Research Complex is a high intensity proton accelerator facility. J-PARC uses high intensity proton beams to create high intensity secondary beams of neutrons, hadrons, and neutrinos [8].
- **PEACER** – Combining Integral Fast Reactor (IFR) approach with the heavy liquid metal cooled reactor technology [9].
- **Thermal Reactors** – Since most commercial reactors are LWR and their spent fuels contain most of the actinides accumulated to date, actinide recycling in LWRs has been extensively evaluated [10].
- **Fast Reactors** – In a fast neutron spectrum, the fission to capture ratio of minor actinides is quite favourable and this is the main reason why there is international consensus that fast reactors are well

suited for minor actinide transmutation. The cross-sections for transmutation reactions are very low, which in turn demands very high neutron fluxes. But fast reactors do have the high neutron fluxes needed. Fast reactors are also considered to be relatively tolerant of large minor actinide loadings because their reactivity control and reactivity feedback parameters are less sensitive than thermal reactors (though this is heavily design dependent and will need to be verified at a later stage of development) [1].

- **Thorium based fuel cycle** – Some Member States are evaluating the thorium fuel cycle as an alternative disposition path for the incineration of plutonium and minor actinides in reactors and as a means for nuclear energy production with low radiotoxicity [10].

- **Inert Matrix Fuel** – Inert matrix fuel concepts have been proposed in order to minimize the number of minor actinide containing fuel pins and maximize the minor actinide transmutation rate, since no additional actinides are generated in minor actinide fuel pins. Both thermal and fast reactor systems are under investigation for utilizing this option [10].

5 Minor Actinide Transmutation in Thermal Reactors

Thermal reactors can achieve worthwhile minor actinide transmutation rates. The balance between neutron fission and captures is less favourable in a thermal reactor and the neutron flux field in thermal reactors is much smaller. However, the thermal neutron cross-sections for minor actinide transmutation are correspondingly larger in a thermal reactor and the transmutation rates are similar. In particular, Am-241 can be transmuted very effectively in a thermal reactor, with up to 70% destroyed within a single irradiation lifetime of a target assembly. The main limitation of thermal reactors is that the total loading of minor actinide targets may need to be restricted to avoid excessive impacts on core reactivity control and reactivity feedback characteristics. Because of such considerations, the potential for minor actinide transmutation in thermal reactors has largely been overlooked in recent years in favour of fast reactors. However, there is potentially an advantage in favour of thermal reactors because there are already more than four hundred in commercial operation, whereas there are onlyfew fast reactor prototypes available. If the international community was to become serious about commercial scale implementation, thermal reactors would be available immediately, whereas a long delay could be expected before the same could be said for fast reactors. Thermal reactors might therefore be useful for early demonstration of minor actinide transmutation on commercially relevant scales, without the delays and uncertainties of having to wait for fast reactor deployment [1].

6 Experimental Results

Minor actinides transmutation indifferent type of thermal reactors is examined. For this purpose, two models of recycling are reviewed: single or one-staged recycling and multiple, or two-staged recycling. The one-staged recycling scheme represents single recycling of minor actinides, gained from spent nuclear fuel in one fuel cycle. E.g. BWR $(10 \times 10 - 8)$ in BWR MOX $(9 \times 9 - 9)$ model represents recycling of minor actinides produced in BWR (10×10–8) spent fuel and recycled in BWR MOX (9×9–9) in only one fuel cycle. After the end of the fuel cycle, which is the only cycle, the residual actinide mass goes for underground storage. The strategy for single recycling of minor actinides leads to limited reduction of minor actinides and the significant production costs remains incompletely compensated.

Multi-stage recycling schemes examined in the report representtwo-staged minor actinides recycling scheme lasting over two nuclear fuel cycles. E.g. VVER-1000 in BWR $(10 \times 10 - 8)$ model is recycling of minor actinides produced in VVER-1000 spent nuclear fuel and recycled in BWR $(10 - 10 - 8)$ in two fuel cycles. All models with only one reactor name in the models signage (from the figures below) as VVER-1000, PWR (w15x15), CANDU and BWR $(10 \times 10 - 8)$ represents recycling of minor actinides produced in the spent nuclear fuels of these reactors and recycled in the same reactors respectively. The two-staged recycling scheme has the potential to reach very low levels of radiotoxicity and heat output as the reasonable numbers of cycles that can be implemented in the fuel cycle are no more than two or three.

6.1 Americium-241 transmutation potential

In Figure 1 it is shown the americium transmutation ratio in the studied reactors. It can be seen that in all type of reactors, except these with the MOX fuel, there is significant transmutation rate of americium-241 up to 85%. The data is in agreement with the current knowledge for the transmutation potential of thermal reactors. The reason for lack of americium-241 transmutation in MOX fuel reactor variants is the high initial plutonium quantity in the startup fuel. Therefore more americium-241 is produced than it is transmuted.

Because Am-241 decays to Np-237, the radiotoxicity due to Np-237 is reduced by about 50% when Am-241 is transmuted to other nuclides upon irradiation in a thermal reactor [2]. Secondly, the radiotoxicity of an americium sample can be reduced with a factor of 10 to 20 after irradiation for 3 to 6 years in a thermal neutron flux of 10^{14} $cm^{-2}s^{1}$. Kloosterman et al., 1994 [2] shows that after a storage time of about 100 years, the radiotoxicity of an irradiated americium sample is considerably reduced compared to the radiotoxicity of an unirradiated sample. The same holds for the alpha activity and the heat emission.

6.2 Neptunium-237 transmutation potential

In regard to the other important minor actinide, neptunium-237, the one-staged recycling reactor schemes

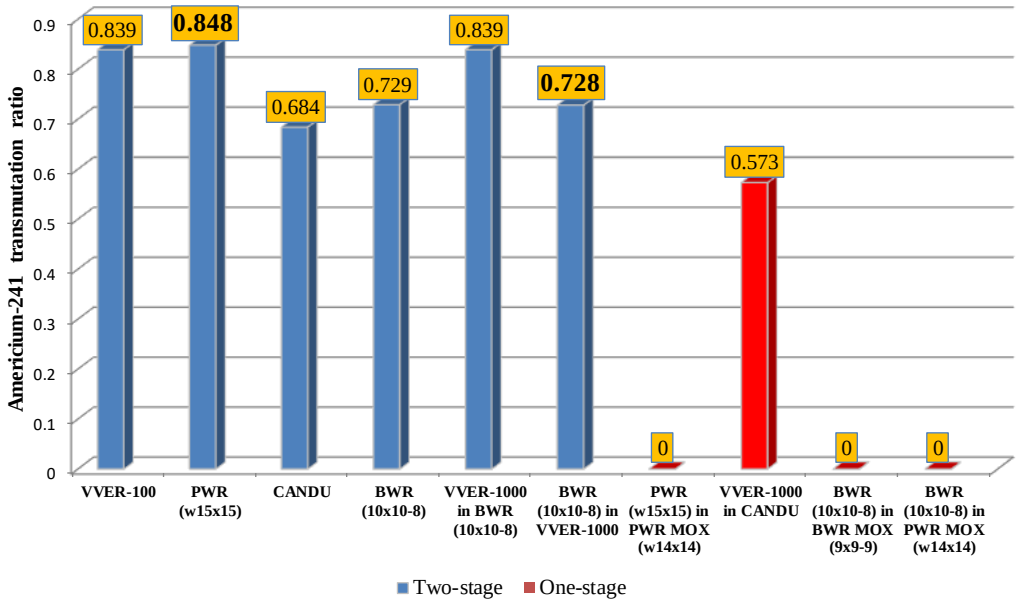

Figure 1. Americium transmutation ratio in different transmutation schemes.

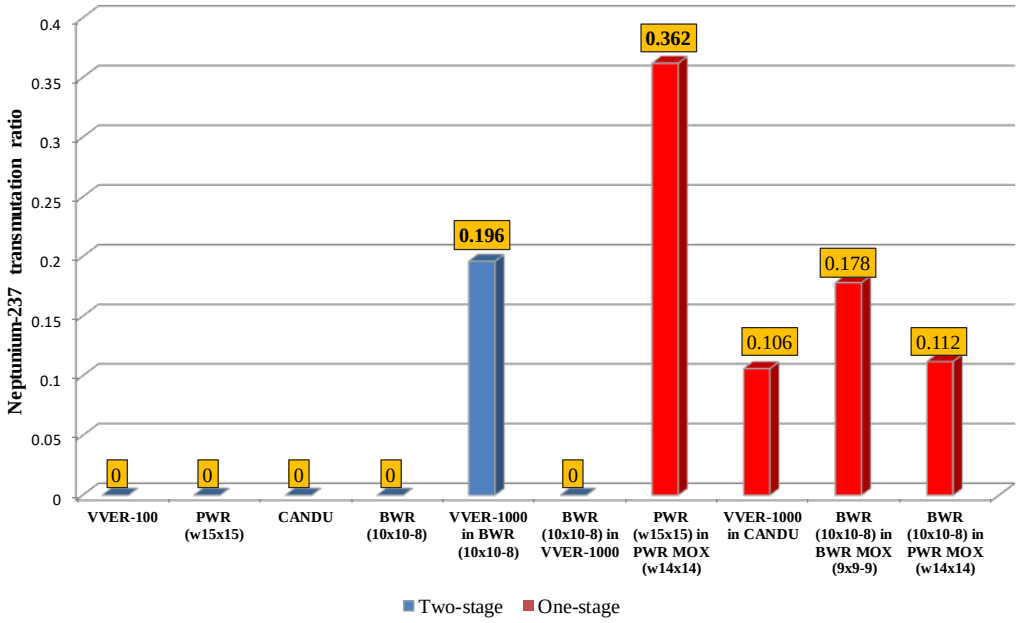

Figure 2. Neptunium transmutation ratio in different transmutation schemes.

with MOX fuel are showing better results than the two-staged transmutation schemes (Figure 2). The reason for which is that neptunium-237 generation is practically not existing in MOX fuels because of the low concentration levels of uranium-235 in the startup fuel (it is depleted uranium that is used). From all two-staged transmutation schemes *only VVER-1000 in BWR* $(10\times10-8)$ shows 20% reduction of neptunium-237 quantity.

6.3 Average Americium-Neptunium transmutation potential

In Figure 3 are shown the average Am-241/Np-237 transmutation ratios in the one and two-staged transmutation schemes. From the figure it is clear that highest transmutation ratio from the all one-staged options possess *VVER-1000 in BWR GE* $(10\times10-8)$ model. The americium/neptunium transmutation potential of this model exceeds 50%. Among the one-staged transmutation schemes, the highest Am-241/Np-237 transmutation capabilities possess *VVER-1000 in CANDU* model. This is also the only one-staged transmutation model with positive ratio.

These results show that thermal reactors with uranium fuel are great transmuters of two of the most concerned, in terms of radiotoxicity, minor actinides: americium-241 and neptunium-237. Moreover, as it was stated above,the americium-241 transmutation results in decreased quantity of generated neptunium-237 in spent nuclear fuel, which reduces the spent nuclear fuel radiotoxicity in long-term scale.

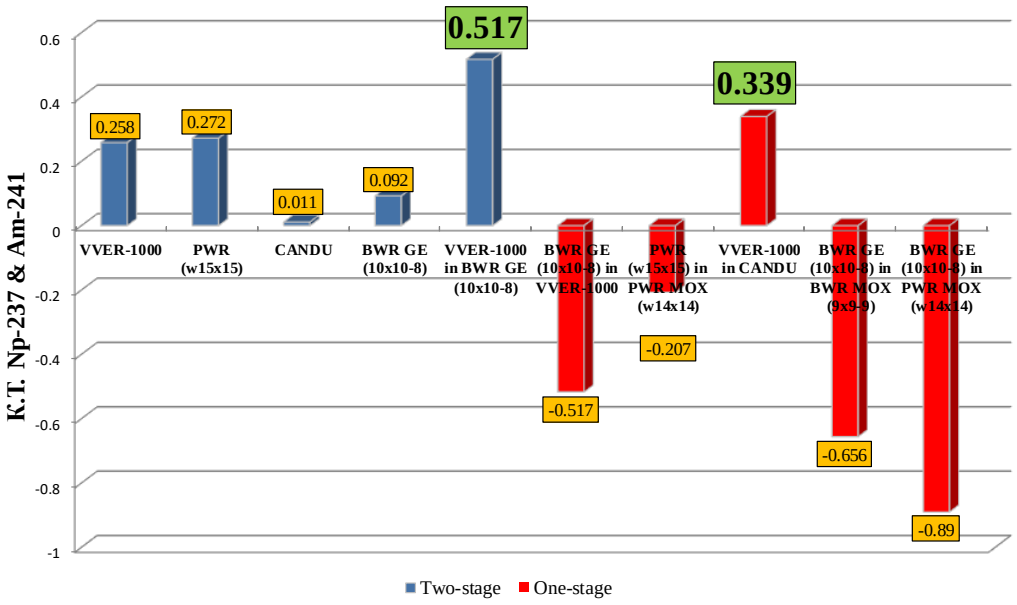

Figure 3. Neptunium-Americium average transmutation ratio in different transmutation schemes.

6.4 Minor actinides transmutation potential

Figure 4 shows minor actinide nuclide concentration change after various recycling schemes. From all two-staged recycling models, the only one with reduction of minor actinide content, compared to the initially charged quantity, is *VVER-1000 in BWR GE* ($10 \times 10 - 8$) model. Charged amount of minor actinides for the first transmutation stage in BWR GE ($10\times10-8$) type of reactors is 1609 gr/tTM. After the second transmutation stage, the minor actinide content is 1312 gr/tTM, which is 18.5% decrease compared to the initially loaded amount. All other two-stage transmutation models shows around 50% average increase of the initially charged minor actinide content.

For all single-stage recycling models, there was no overall reduction in minor actinides content after the end of the recycling stages. However, the model of *VVER-1000* spent nuclear fuel recycling in *CANDU* type of reactors shows most insignificant increase of the amount of minor actinides after the recycling stages. Minor actinides quantity before the first transmutation stage is 1609 gr/tTM, but after the end of the recycling scheme in the CANDU type of reactors, there is insignificant 1.9% increase, which is equivalent to 1640 gr/tTM, or only 31 gr/tTM differences. All other one-staged transmutation models increase more than two times their initially charged minor actinide content with the highest value in possession on BWR GE ($10\times 10-8$) in PWR MOX ($\mathrm{w}14\times14$) model.

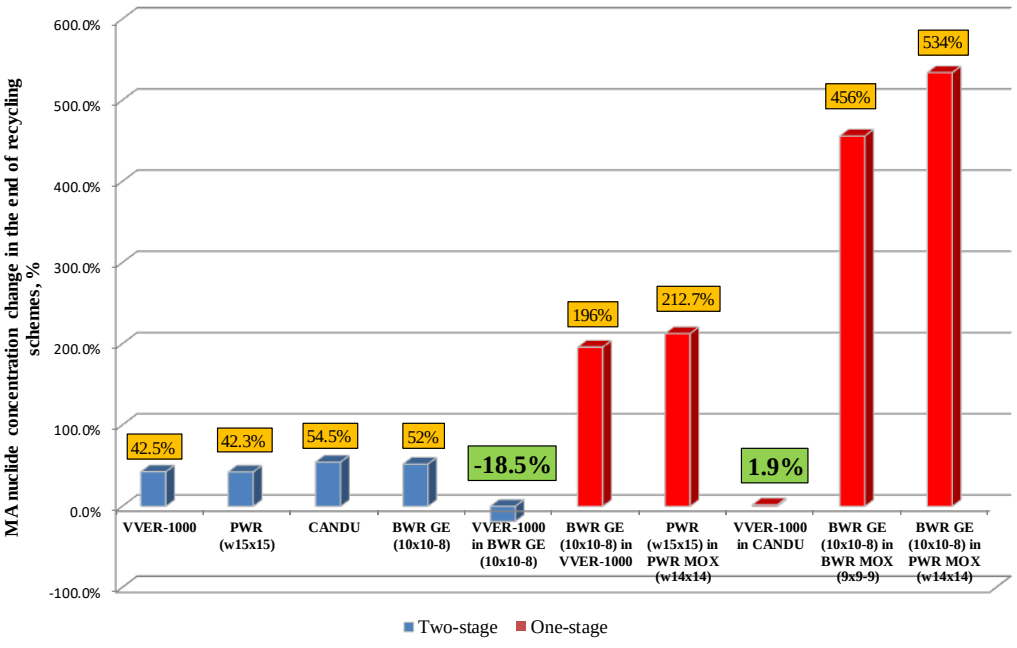

Figure 4. Minor actinide nuclide concentration change in different transmutation schemes.

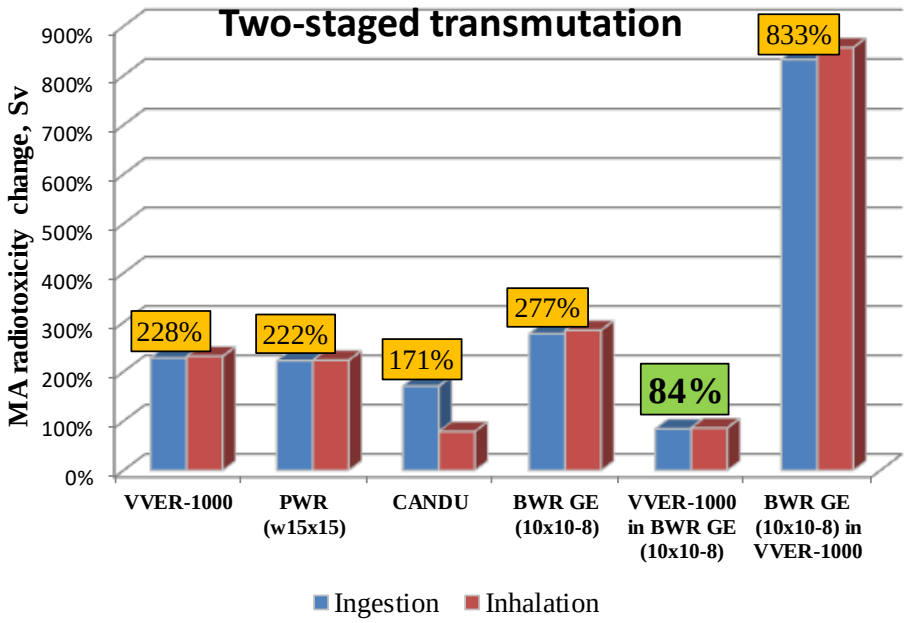

Figure 5. Minor actinides radiotoxicity change in two-stage recycling schemes.

6.5 Minor actinides radiotoxicity change

Radiotoxicity and gamma decay heat are two main characteristics of spent nuclear fuel that define not only the way storage of fuel after its removal from the reactor core, but the long-term danger, which they pose to human and environment [11]. The decay heat of radioactive waste defines the basic parameters of the systems for heat removal which serve spent nuclear fuel in at least five years after its removal from the reactor and radiotoxicity determined radiological hazard of radioactive waste after its leakage and penetration into the environment.

In Figure 5 it is shown the percentage change of radiotoxicity due to minor actinides share in spent nuclear fuel on the two-staged transmutation models. All models show significant increase of the minor actinides radiotoxicity except the *VVER-1000 in BWR GE* ($10\times10-8$) variant. The minor actinide radiotoxicity after two-staged transmutation cycles in BWR reactors is only 84% more than the minor actinide radiotoxicity of the startup fuel. It is visible also that despite of the **-18.5%** decrease of minor actinide nuclide concentration content in the end of the transmutation scheme, the radiotoxicity in this model is increased, not decreased. The reason for this is the type of the transmutation of americium-241 which is related with the lower energy spectrum of the studied thermal reactors. Because it is not fissible, it is transmuted by neutron capture to Am-242m (10%) or to Am-242 (90%). The first-mentioned activation product has a relatively long half life of 141 years, is highly fissile, and can therefore easily be fissioned in a

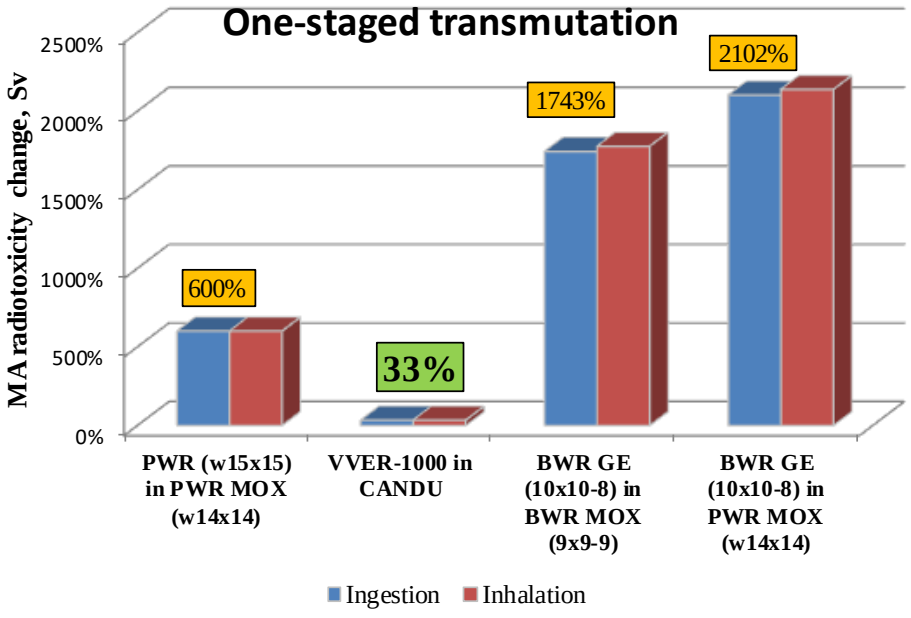

Figure 6. Minor actinides radiotoxicity change in one-stage recycling schemes.

thermal neutron flux. The second activation product also has a high fission cross section, but decays with a half life of 16 hours to Cm-242 (83%) or Pu-242 (17%). Because of this type of transmutation through neutron capture, high mass radiotoxic actinides suchas Cm-243, Cm-244, Cm-245 are produced and the excess radiotoxicity can not be compensate with the reduced minor actinides concentration.

In Figure 6 there is shown the percentage change of radiotoxicity due to minor actinides share in spent nuclear fuel in the one-staged transmutation models. All one-stage models show significant increase of the minor actinides concentration, up to 21 times more, as in *BWR GE (*$10 \times 10 - 8$*) in PWR MOX (w14x14)*, compared to the initial startup quantity. Only *VVER-1000 in CANDU* model show 33% insignificant increase of the minor actinide content. The reason for this is again lack of plutonium in the startup fuel and therefore lower americium generation through the transmutation cycle.

7 Discussion

Minor actinides transmutation in thermal reactors covers two of the three main objective of the partitioning and transmutation strategy of actinides:

1. *Reduction of the total radiotoxic inventory of underground disposal sites;*
2. *Using fissile actinides for energy production.*

VVER-1000 in BWR GE ($10 \times 10 - 8$*)* model shows that thermal reactors can be very good transmuters when high initial quantities of minor actinides are loaded with the startup fuel. All of the experimental studies for minor actinides transmutation in thermal reactors confirm that worthwhile quantities can be recycled in LWRs, with useful quantities destroyed in the irradiation lifetime of the fuel assemblies containing the minor actinides [12]. The majority of commercial reactors in operation in theworld are thermal reactors, and if we get the minor actinides transmuted in the thermal reactors during their power generating,the inventory of high level long-lived radioactive minor actinides in the world will be greatly reduced.

In terms of minor actinides share in the total energy production, their impact on thermal-hydraulic reactor characteristics is widely analyzed. All of the studies [12] confirm satisfactory core and thermal-hydraulic behaviour during minor actinides transmutation cycles. The fact that so many independent studies have reached the same conclusion is also very strong evidence of feasibility.

In regard to reduction of radiological dose to the population and environment, minor actinides transmutation in thermal reactors shows radiotoxicity increase of the minor actinides in spent nuclear fuel, especially of the MOX fuel models. Therefore, minor actinides transmutation is not the best option in regard to reduction of the possible dose to the population due to leakage of actinides from underground disposal sites.

8 Conclusions

Minor actinides transmutation in thermal reactors represents excellent option for reducing the total radiotoxic inventory of underground disposal sites. During the recycling cycles, there is satisfactory thermal-hydraulic core behaviour, which is another benefit of actinides transmutation in thermal reactors. Minor actinides transmutation in thermal reactors could have also play a role as a technology demonstrator for more advanced recycling schemes that might follow in the future, when Generation IV reactor systems are operational [12]. Last but not least, minor actinides transmutation in thermal reactors results in enhanced proliferation resistance of the spent nuclear fuels, because of drastically increased plutonium-238/plutonium ratio.

References

[1] Minor Actinide Transmutation. (accessed ...). Retrieved from National Nuclear Laboratory.co.uk: http://www.nnl.co.uk/media/1053/minor_actinide_transmutation_-_position_paper_-_final_for_web1.pdf.

[2] Kloosterman J.L., De Kruijf W.M. (1994) Transmutation of Americium in Thermal Reactors. Cadarache, France: Third International Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation.

[3] Adlys G. (accessed ...). Radiotoxicity Dynamics in Fission Products. Department of Physics, Kaunas University of Technology.

[4] INTERNATIONAL ATOMIC ENERGY AGENCY (2004) Application of the Concepts of Exclusion, Exemption and Clearance.

[5] Paveslescu A., Gepraga D. (2007) CANDU Radiotoxicity Inventories Estimation – A Calculated Experiment Cross-check For Data Verification and Validation. *Rom. J. Phys.* **52** 137–148.

[6] Murata I., et al. (2007) New burnup calculation system for fusion–fission hybrid reactor. *Fusion Engineering and Design* **82** 2772-2778.

[7] Fredberg J.P., Kadak A.C. (2009) Fusion–fission hybrids revisited. *Nature Physics* **5** 370-372.

[8] Sasa T. (2013) Multipurpose application of the J-PARC Transmutation Experimental Facility. Presented at 10th ASRC International Workshop on Nuclear Fission and Decay of Exotic Nuclei, March 21–22, Tokai, Japan.

[9] Park B.-G., Jeong S.-H., Hwang I.-S. (accessed ...) Partitioning and transmutation of spent nuclear fuel by PEACER. Seoul National University.

[10] INTERNATIONAL ATOMIC ENERGY AGENCY (2009) Status of Minor Actinide Fuel Development.

[11] Bergelson B.R., Gerasimov A.S., Tikhomirov G.V. (2005). Radiotoxicity and decay heat power of spent nuclear fuel of VVER type reactors at long-term storage. *Radiat. Prot. Dosimetry* **115** 445-447.

[12] OECD-NEA (2013) Minor Actinide Burning in Thermal Reactors. A Report by the Working Party on Scientific Issues of Reactor Systems.