Investigating material attractiveness of minor actinides fuel for waste transmutation

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Abstract

Radiotoxicity of nuclear waste is dominated by fission products in the first ~500 years, while for longer times long-lived transuranic elements play a larger role. The main purpose of partitioning and transmutation (P&T) is to transform nuclides that represent a long-term source of heat and radiotoxicity, such as plutonium and minor actinides (MAs) into stable and short-lived nuclides. The use of nitride fuel for fast reactors has gained attention because besides its higher melting point and thermal conductivity compared to oxide fuels. Additionally, actinide mononitride can accommodate a wide range of MA compositions, enabling nitride fuels to be used for transmutation in fast reactors. However, studies of non-proliferation implications of using such fuel are scarce. In this paper we present results related to investigation of material attractiveness plutonium produced during irradiation of americium-neptunium nitride fuel, intended to fuel a lead-cooled reactor. The plutonium content in the spent MA nitride fuel was evaluated in light of material properties known to impact the usability of the material in the manufacture of nuclear explosive devices. The material attractiveness evaluation was done using simulations and material attractiveness figures-of-merit suggested in earlier works.

1. Introduction

Nuclear waste transmutation via fissioning is an option to tackle the high long term radiotoxicity and heat from fission products in spent fuel. In the first 30 years of cooling time, the major contribution to radiotoxicity is caused by fission products, such as ^{85}Kr, ^{90}Sr ($T_{1/2}$ = 29 years) and ^{137}Cs ($T_{1/2}$ = 30 years). While the radiotoxicity of fission products decreases significantly after 1,000 years, the transuranic inventory in spent fuel remains highly radiotoxic for more than 100,000 years. The main isotopes that contribute to this effect are ^{239}Pu ($T_{1/2}$ = 24,112 years), ^{240}Pu ($T_{1/2}$ = 6,563 years), ^{241}Am ($T_{1/2}$ = 432 years) and ^{243}Am ($T_{1/2}$ = 7.370 years) [1].

Actinides, such as plutonium (Pu) and americium (Am) can be transmuted to short lived fission products, which have reduced toxicity and volume. Commercial recovery of plutonium and uranium from Spent Fuel (SF) for use in mixed oxide fuel (MOX) has already been performed by some states. In addition, options to decrease the quantities of minor actinides (MA), i.e. Cm, Am, Np, in the waste are also of interest [9].

The use of MA fuel or targets in fast reactors have been investigated over the years [2-8], and the crystal structure of transuranic nitrides seems to present advantages over the oxide option [9], such as higher melting point and thermal conductivity.

Wallenius [10] proposes a compact lead cooled reactor design to perform MA transmutation using (Np,Am)N fuel. The fresh fuel would be composed only of Neptunium (Np) and Americium (Am), summing up 373 kg (32wt.%^{237}Np, 58wt.%^{241}Am, 10wt.%^{243}Am) for one reactor core unit. This means that large quantities of MA, over 100kg, would be incorporated into the fuel cycle of the proposed reactor design. This would be the case first in the

beginning of the fuel cycle, during the fuel fabrication process, and maybe in the end of the cycle, since still a big quantity of MA is expected to be found in the spent fuel.
Although plutonium and uranium (HEU and ^{233}U) are under international safeguards, MAs, such as Np and Am are monitored by the IAEA international safeguards exclusively under voluntary arrangements with States [11]. Despite that, some isotopes have bare critical masses comparable to uranium or plutonium. In this work, the fuel material intended for the lead-cooled fast reactor described in [10] was selected for material attractiveness analysis, as it may constitute a challenge from a non-proliferation perspective. The material attractiveness analysis was done by assessing parameters impacting the material's usability in a nuclear explosive device as well as barriers against its use.

2. Methodology

2.1 Reactor design and life-cycle

The lead-cooled reactor core design and fuel, developed in [10], were used as the basis for the analysis. The main features of the lead-cooled reactor core are presented in Table 1. The irradiation of the fresh fuel was modeled in the Monte Carlo code Serpent2 [12], assuming irradiation during the reactor life-time of 24 years without refueling, which when considering an availability of 90% corresponds to 8000 full power days. A power density of 15 W/g of actinides was assumed.

Table 1: Main geometry parameters of the reactor technology considered in this paper (For more details, see [10]).

Core Design [10]			
	Number of assemblies	rods/ assembly	material
fuel assemblies	7	61 rods	(Np,Am)N
control assemblies	6	19 rods	B_4C
shutdown assemblies	6	7 rods	$(W,Re)^{10}B_2$
reflector assemblies	18	37 rods	YSZ - yttria-stabilised zirconia
Fuel Rod [10]			
fuel pellet	diameter	13.4 mm	
	column height	540.0 mm	
	fuel material	(Np,Am)N (32wt.%^{237}Np, 58wt.%^{241}Am, 10wt.%^{243}Am)	
cladding	thickness	0.5 mm	

	material	alumina scale material [13, 14] .

2.2 Attractiveness Parameters Calculation

Many authors [15-21] evaluate attractiveness of nuclear material to be used in a nuclear explosive device based on the following parameters: bare critical mass, radiation dose rate, decay heat and neutron emission rate. Each parameter will now be described along with the computational methodology used to compute it:

Bare Critical Mass (BCM): The bare critical mass is the minimum amount of the material required to sustain a chain reaction without initiator or reflector. Therefore, materials with smaller BCM are preferred for a nuclear explosive device manufacture. Serpent2 code was used to calculate the minimal radius of a sphere constituted of the desired material that generates a criticality parameter, $k_{eff} \geq 1.000$ through linear interpolation. The cross section library used in Serpent was JEF-3.1.1 [22], and the temperature of the material considered was 300 K.

Radiation Dose Rate: The radiation dose is evaluated because it is related to the management of the nuclear material and also because it may impact the high explosives [19]. A low radiation dose increases the attractivity of the material. The dose was calculated separately for gammas and neutrons via MCNP 6.2 [23] with the tally F2, using the photon dose function from ICRP-21 1971 and the ICRP-60 effective-dose conversion function for energy deposition tally.

Decay Heat: Decay heat represents heat generated by radioactive decays in the nuclear material and may cause difficulties with respect to handling of the material as well as with respect to the high explosives. A low decay heat makes the material more attractive for a potential proliferator [24]. The decay heat generated by individual nuclides was calculated via Serpent2 code.

Neutron Emission Rate: The spontaneous fission neutron rate can affect the yield and reliability of a nuclear explosive device. For instance, a high spontaneous fission rate can lead to a premature detonation producing a lower yield or a so-called fizzle [25]. The spontaneous fission rate generated by each nuclide was calculated via Serpent2 code and the multiplicities for different nuclides considered for the calculation of the neutron emission rate.

In order to quantify the attractiveness of the material, the two figures of merit (FOM) proposed by Bathke *et. al.* [15] were calculated. It is assumed that the nuclear material acquired after reprocessing has taken place, and two different groups of proliferators are analyzed. For the first group neutron emission rate is not considered an issue, described by [15] as subnational groups and technically advanced proliferant states, the former because they are satisfied with lower yield and the latter because they can overcome the issue while maintaining a high yield. For this first group FOM_1 is calculated (Eq. 1), taking into

consideration BCM, dose rate and heat decay rate. For the second group, neutron production rate is considered an issue, described by [15] as relatively unadvanced proliferant states. For this second group a term related to neutron emission rate is added to the calculation of FOM_2 (Eq. 2).
The equation for the two different FOMs are presented:

$$FOM_1 = 1 - log_{10}\left(\frac{BCM}{800} + \frac{BCMh}{4500} + \frac{BCM}{50}\left[\frac{D}{500}\right]^{1/log_{10}2}\right) \quad \textbf{(1)}$$

$$FOM_2 = 1 - log_{10}\left(\frac{BCM}{800} + \frac{BCMh}{4500} + \frac{BCMS}{6.8(10)^6} + \frac{BCM}{50}\left[\frac{D}{500}\right]^{1/log_{10}2}\right) \quad \textbf{(2)}$$

where,

BCM = bare critical mass of the metal (kg); h = decay heat (W/kg); D = dose rate evaluated at 1m from the surface (rad/h); S= spontaneous fission neutron rate (n/s.kg-1).

The values calculated for the FOM_1 and FOM_2 have the following interpretation according to [15]: A FOM value smaller than 1 corresponds to a material that is considered unattractive for use in a nuclear explosive device; A FOM value between 1 and 2 means that the material is considered attractive; and FOM > 2 means that the material is considered preferred to be used in a nuclear explosive device.

2.3 Actinides production pathways

In order to understand the nuclide vector that is found after irradiation of the nuclear fuel, the main paths for transmutation and decay processes along with their half-lifes ($T_{1/2}$) are presented in Fig.1 and 2.

2.3.1 Neptunium

The fresh fuel core consists of 120.25 kg of Neptunium. After 24 years of irradiation, the core is expected to still consist of large quantities of neptunium although a fraction has undergone transmutation.During irradiation, the first transmutation product of ^{237}Np is ^{238}Np, which has a very short half-life compared to the lifecycle time of the reactor core. ^{238}Np may in turn absorb a neutron and produce ^{239}Np, or undergo beta decay to produce ^{238}Pu which in turn can absorb a neutron and create ^{239}Pu, or even ^{240}Pu after absorption of yet another neutron, see Fig 1.

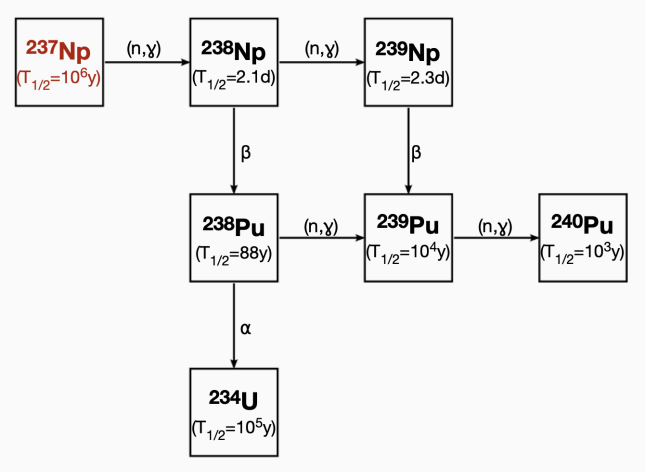

Fig. 1: Main transmutation and decay paths derived from the ^{237}Np in the (Np,Am)N fresh fuel, along with their half-lifes ($T_{1/2}$).

2.3.2 Americium

^{241}Am produces ^{242}Am and ^{242m}Am via neutron absorption, see Fig 2. ^{242m}Am decays to ^{242}Am via isomeric transition (IT) with a half-life of 141 years. Due to the large quantities of ^{241}Am in the fresh fuel and the higher probability (> 35%) for its metestate formation, especially for high energy neutrons (E_n> 30 keV) [26], a higher content of ^{242m}Am is expected in spent fuel. Although ^{242m}Am has as low heat production (~4.2W/kg [15]) compared to ^{242}Am (~9x10^{5}W/h [15]), the separation of its metastable isotope from other isotopes is very difficult, which can make its use very difficult. ^{242}Cm builds up as ^{242}Am beta-decays with a half-life of only 16 h. ^{242}Cm subsequently decays via alpha decay to ^{238}Pu with a half-life of 163 days. ^{238}Pu alpha-decays into ^{234}U, which is more stable with half-life in the order of 10^5 years.
^{244}Am is produced from ^{243}Am via neutron absorption, see Fig 2. ^{244}Am beta-decays to ^{244}Cm, which can also absorb a neutron and generate ^{245}Cm. ^{240}Pu is built up by the decay of ^{244}Cm via alpha-decay.

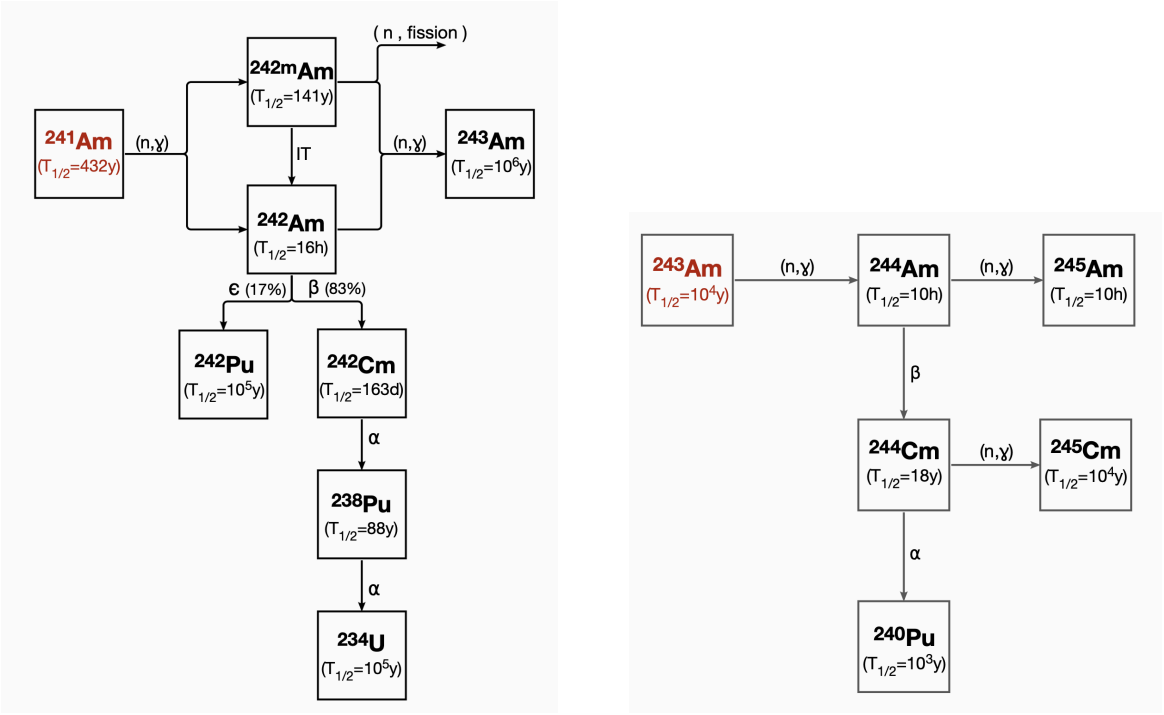

Fig.2: Main transmutation and decay paths derived from the americium isotopes in the (Np,Am)N fresh fuel, along with their half-lifes ($T_{1/2}$).

2.3.3 Plutonium

At this point we can notice that the ^{238}Pu isotope would be formed in the reactor in quantities proportional to ^{237}Np and ^{241}Am transmutation rate. Other plutonium isotopes, such as ^{239}Pu and ^{240}Pu will also build up during the reactor life cycle. The quantities of ^{238}Pu might be

significantly reduced 100 years after irradiation, while other plutonium isotopes won't present a significant decrease, due to their very long half-lifes.

The study of separated isotopic elements of the material of interest for this research was already done by Bathke *et al.* [15], therefore in our study we focus on the element vectors that include all the isotopes of the element of interest.

3. Results and Discussion

Uranium and plutonium must be analyzed due to its importance to safeguard agreements. We also propose that the MA, neptunium, americium and curium deserve a material attractiveness analysis, since the existence of isotopes that can represent a proliferation concern. Due to the limited number of pages in the current publication, only the results related to plutonium vectors in spent fuel will be presented in the results section.
The attractiveness analysis of the plutonium content in (Np,Am)N fuel was done taking into account the following considerations:

- There is a reprocessing step at the end of the fuel cycle to recover plutonium that can take place in different stages: the reactor end of lifecycle (after 24 years of operation) or after being in an interim storage for 2, 5, 10, 30, 60 or 100 years of cooling time (CT). This choice of time is mainly to verify how this fuel would evolve during the years compared to more conventional nuclear fuel and to check for any particular moment where the plutonium content would be more attractive;
- The plutonium vector in analysis is considered in the form of metal for the calculation of the bare critical mass and dose rate.

3.1 Plutonium isotopic composition

The plutonium composition and masses for the selected cooling times are presented in Table 2.

Table 2: Plutonium vector total mass and weight fractions for different cooling times (CT).

CT (years)	total mass /assembly (kg)	Pu isotopes weight fraction						
		236	238	239	240	241	242	244
0	5.202	3.196×10^{-7}	0.814	0.022	0.039	5.871×10^{-4}	0.124	4.987×10^{-5}
2	5.256	1.952×10^{-7}	0.810	0.022	0.045	5.281×10^{-4}	0.123	4.936×10^{-5}
5	5.207	9.568×10^{-8}	0.799	0.022	0.053	4.619×10^{-4}	0.124	4.982×10^{-5}
10	5.115	2.962×10^{-8}	0.783	0.023	0.066	3.705×10^{-4}	0.126	5.072×10^{-5}
30	4.712	1.212×10^{-9}	0.730	0.027	0.104	1.570×10^{-4}	0.138	5.506×10^{-5}
60	4.115	1.107×10^{-9}	0.665	0.033	0.141	4.791×10^{-5}	0.159	6.305×10^{-5}

CT (years)	total mass /assembly (kg)	Pu isotopes weight fraction						
		236	238	239	240	241	242	244
100	3.470	1.321×10^{-9}	0.585	0.045	0.177	1.589×10^{-5}	0.191	7.519×10^{-5}

The major plutonium isotope in spent fuel is ^{238}Pu, followed by ^{242}Pu and ^{240}Pu. Because of its short half-life. ^{238}Pu content decreases with increasing cooling time, and only 47% remains after 100 years. ^{240}Pu instead increases with increasing cooling time, due to the alpha decay of ^{244}Cm. This leads to a build up of approximately 4.3kg of ^{240}Pu in the full core, corresponding to 17.7% of the total mass of plutonium in spent fuel after 100 years of cooling time.

3.2 Material attractiveness analysis

The parameters: bare critical mass (BCM), radiation dose rate, decay heat and neutron emission rate of the produced plutonium in spent fuel were analyzed as a function of cooling time, Figure 3. The values of BCM, decay heat, dose rate and neutron emission rate vary differently with respect to cooling time. The BCM value increased 30% after 100 years of cooling time, while the other parameters decreased. Decay heat, neutron emission rate and dose decreased 28%, 14% and 27%, respectively. These are mainly related to the decrease in the content of ^{238}Pu.

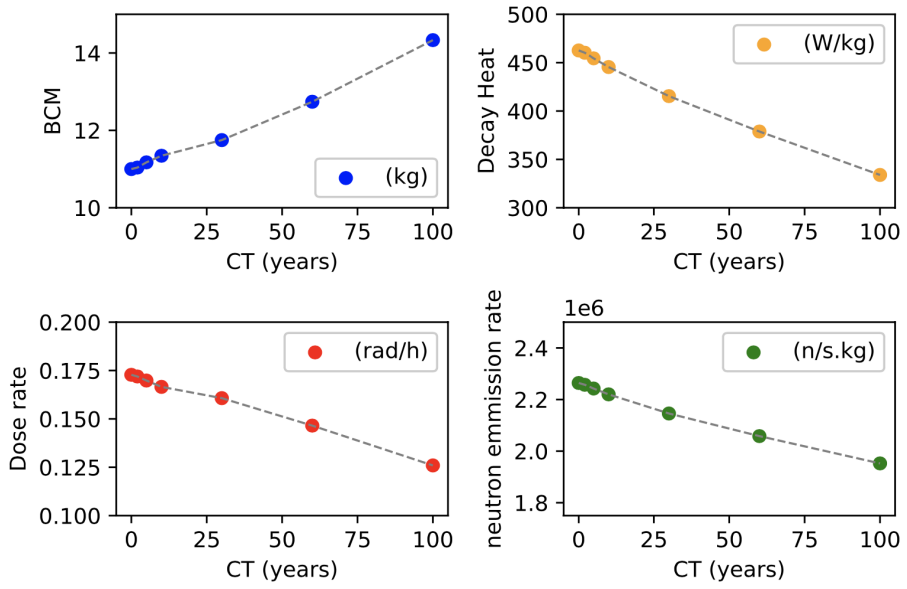

Fig.3: Bare Critical Mass (BCM), decay heat, radiation dose rate and neutron emission rate (SF) of the plutonium vectors as function of the cooling time.

The calculated values for FOM_1 and FOM_2 for the different plutonium compositions are presented in Figure 4, along with FOM_1 and FOM_2 values for the plutonium isotopes ^{238}Pu,

^{240}Pu and ^{242}Pu as reference. The FOM_1 values for the plutonium isotopes used as reference were obtained from Bathke *et. al.* [15], while the FOM_2 values were calculated in this paper.
For the plutonium vectors, no major dependencies of the cooling time were observed for the values of FOM_1 and FOM_2. The FOM_1 values were calculated varying from 0.94 to 0.96 after 100 years of cooling time. The explanation is that the decrease in dose rate and decay heat with increasing cooling time is compensated for by an increasing critical mass. Values of FOM_2 are shown in Fig.4, and are found between 0.32 and 0.28, for 0 and 100 years respectively. A bigger variation appears here, due to the addition of the neutron emission rate parameter in the calculation. Both FOMs values classifies the material as unattractive for the first 100 years of cooling time, according to ref [15].

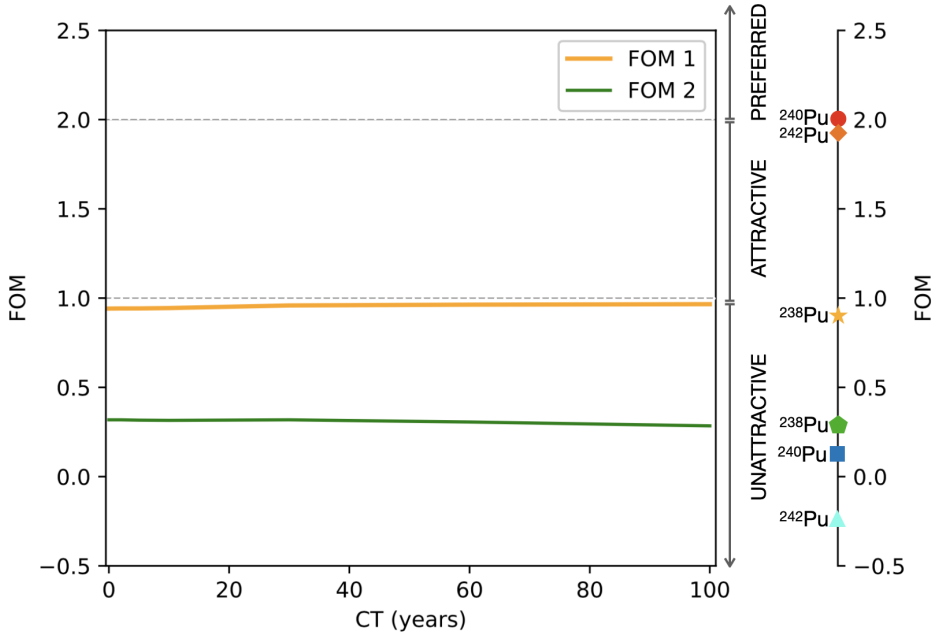

Fig. 4: Figure of merit (FOM) as a function of cooling time for the respective plutonium isotopes vectors. The symbols star, circle and diamond denote FOM_1 values for the pure isotopes of ^{238}Pu, ^{240}Pu and ^{242}Pu respectively [15]. The symbols pentagon, square and triangle denote FOM_2 values for the pure isotopes of ^{238}Pu, ^{240}Pu and ^{242}Pu respectively. Dashed lines were used to delimitate the areas of the plot that that relates the values of FOM to the attractiveness level of the material- i.e., Unattractive for FOM < 1, Attractive for 1 < FOM < 2 and Preferred for FOM > 2.

4. Conclusions

Nuclear waste with long-lived radiotoxic properties is a major concern for society and stakeholders in the nuclear sector. Nuclear waste transmutation via fissioning is an option to tackle the high long term radiotoxicity and heat from fission products in spent fuel.
In this paper we presented initial results related to material attractiveness analysis of the plutonium in spent fuel americium-neptunium nitride, intended to waste transmutation in a lead-cooled reactor.

The depletion of the MA nitride fuel was performed in Serpent2 and from the results, the plutonium vector could be determined. Radiation dose rate, decay heat, neutron emission rate and bare critical mass were calculated with computational simulations. The material attractiveness was then evaluated using two different figures-of-merit suggested by Bathke et al [15].

The plutonium present in the spent nuclear fuel is dominated by ^{238}Pu, composing 81% of the plutonium content at discharge. After 5 years of cooling time, the relative ^{238}Pu content is still close to 80%. It is also valid to highlight that the mass of plutonium produced by the entire reactor core (5.2kg immediately after discharge) is lower than one significant quantity (8.0kg) defined by IAEA [11]. Calculations of the two FOMs revealed that the material is unattractive for all the evaluated cooling times.

The complete set of results - including the evaluation of attractiveness parameters for the vectors of neptunium, curium, americium and uranium, and a combination of plutonium and MA in consideration of advanced reprocessing approaches - will be presented along with nuclear safeguards and security considerations in a separate publication.

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References

1. Wallenius, J., Transmutation of Nuclear Waste, 2011.
2. M. Osak, H. Serizama, M. Kato, K. Nakajima, Y. Tachi, R. Kitamura, S. Miwa, T. Iwai, K. Tanaka, M. Inoue, and Y. Arai, Research and development of minor actinide-containing fuel and target in a future integrated closed cycle system, J. Nucl. Sci. and Tech. 44 (2007), p. 309–316.
3. D.Neuman, C.C.Davis, T.A.Nothwang, F.G.Hampel, S.L.Voit, M.R.Lopez, and A.C.Martinez, Characterization of minor actinide mixed oxide fuel, J. Nucl. Mater 385 (2009), p. 168–172.
4. Status and advances in MOX fuel technology, Tech. Rep. 415, IAEA Technical Reports Series - International Atomic Energy Agency, Vienna, (2003).
5. F. Sudreau, J. Bonnerot, E. Brunon, D. Warin, G. Gaillard-Grol´eas, and M. Ferroud-Plattet, Oxide fuels and targets for transmutation, Tech. Rep. NEA 6282, OECD/NEA's 9th Info. Exchange Mtg of Actinide and Fission Product Partitioning and Transmutation, Paris, (2007).
6. K. Minato, M. Akabori, M. Takano, Y. Arai, K. Nakajima, A. Itoh, and T. Ogawa, Fabrication of nitride fuels for transmutation of minor actinides, J. Nucl. Mater 320 (2009), p. 18–24.
7. Y.A. et al., Development of nitride fuel and pyrochemical process for transmutation of minor actinides, Tech. Rep. NEA 6282, OECD/NEA's 10th Info. Exch. Mtg on Actinide and Fission Product Partitioning and Transmutation, Paris (2008).
8. M.Streit, F.Ingold, M.Pouchon, L.J.Gauckler, and J.-P.Ottavianic, Zirconium nitride as inert matrix for fast systems, J. Nucl. Mater 319 (2003), p. 51–58
9. Implications of partitioning and transmutation in radioactive waste management. — Vienna : International Atomic Energy Agency, 2004. Technical reports series, ISSN 0074–1914 ; no. 435.

10. J.Wallenius, Maximum efficiency nuclear waste transmutation, Annals of Nuclear Energy 125 (2019), p. 74-79.
11. IAEA Safeguards Glossary, Tech. Rep., International Atomic Energy Agency, Vienna, (2002).
12. J. Leppänen, M. Pusa, T. Viitanen, V. Valtavirta, and T. Kaltiaisenaho. *"The Serpent Monte Carlo code: Status, development and applications in 2013."* Ann. Nucl. Energy, 82 (2015) p. 142-150.
13. J. Ejenstam, M. Halvarsson, J.W.B. Jonsson, and P. Szakalos Oxidation studies offe10cral-re alloys exposed to pb at 550c for 10000 h, Journal of Nuclear Materials 443 (2013), p. 161–170.
14. J. Ejenstam and P. Szakalos, Long term corrosion resistance of alumina forming austenitic stainless steels in liquid lead, Journal of Nuclear Materials 461 (2015), p. 164–170.
15. C. G. Bathke, et al., "The attractiveness of materials in advanced nuclear fuel cycles for various proliferation and theft scenarios", Nuclear Technology, Vol.19 (2012).
16. J. Magil, "Non-proliferation and Safeguards Activities at ITU - Recent Publications", Technical Report, JRC-ITU-TN-2000/19 (2000).
17. D.E.Beller and R.A. Krakowiski, Burn-up dependance of proliferation attributes of plutonium from spent LWR fuel, LA-UR-99-751, Los Alamos Laboratory (1999).
18. B. B. Ebbinghaus, C. G. Bathke, D. S. Dalton, J. P. Murphy, The Application of Material Attractiveness in a Graded Approach to Nuclear Materials Security, LLNL-CONF-640708 (2013).
19. Pellaud B, Proliferation aspects of Plutonium Recycling, J. Nucl. Mat. Management, 31, 30 (2002).
20. Sagara, H, et al., Denaturing of Plutonium by Transmutation of Minor-Actinides for Enhancement of Proliferation Resistance, J. Nucl. Sci. Technology, 42, 161 (2005).
21. Saito, M., et al, Development of Methodology for Plutonium Categorization (II) - Improvement of Evaluation Function 'Attractiveness', American Nuclear Society Transactions, 98, 1, 669 (2008).
22. A. Santamarina, et al. The JEFF-3.1.1 Nuclear Data Library, JEFF Report 22, OECD/NEA No. 6807 (2009).
23. *MCNP® USER'S MANUAL Code Version 6.2* , Los Alamos National Laboratory report LA-UR-17-29981
24. IAEA-SM-367/15/07, PROLIFERATION-RESISTANCE AND SAFEGUARDABILITY OF INNOVATIVE NUCLEAR FUEL CYCLES.
25. *Evaluation of Proliferation Resistance of Plutonium Based on Decay Heat.* Kimura, Yoshiki, Saito, Masaki and Sagara, Hiroshi. 5, Journal of Nuclear Science and Technology, Vol. 48 (2011), p. 715-723.
26. K. Wisshak, J. Wickenhauser, F. Käppeler, G. Reffo & F. Fabbri, The Isomeric Ratio in Thermal and Fast Neutron Capture of Americium-241, Nuclear Science and Engineering, 81:3, 396-417, DOI: 10.13182/NSE82-6 (1982) .