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Change of the Quality of the Material Aspects of the Intrinsic Proliferation Barrier of Reactor-Grade Plutonium Produced in a PWR as a Function of Fuel Type, Cooling Time, and Burn-Up

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Abstract. In the recent years, non-proliferation of nuclear materials has become an important issue, regarding civilian fuel cycle's development. In addition, future nuclear energy systems' and nuclear fuel cycles' advancement should comply with non-proliferation criteria. These requirements derive from the circumstance that nuclear power is considered to be a dual purpose technology. Special attention to proliferation resistance has been paid since 2000, the main concerns arising from plutonium stockpiles accumulation, and plutonium being considered among the most important materials related to non-proliferation. There is an on-going debate about reactor-grade plutonium's usability for nuclear explosives manufacturing. Furthermore, there isn't universally recognized and verified methodology for proliferation resistance assessment. On the other hand, intrinsic material factors may play further role in strengthening fuel cycles' proliferation resistance, since the majority of the efforts in that area has gone so far into enhancing technical barriers. The article presents an analysis of the change of the intrinsic barrier's quality of reactor-grade PWR plutonium as a function of fuel type, burn-up, and cooling time, the main focus being on the physical components. The assessment has been carried out using three different methodologies in an attempt to evaluate their applicability in intrinsic material barrier analysis.

Keywords: material barrier, proliferation resistance, reactor-grade plutonium.

1 Introduction

The relevance of the problems, associated with nuclear materials' non-proliferation is defined by the following circumstances: (1) nuclear power is considered to be a dual purpose technology [1, 2]; and (2) the probability for proliferation creates certain obstacles before nuclear power's development in terms of creating negative perception of nuclear technology [3]. On the other hand, proliferation resistance is defined as a main requirement for Generation IV nuclear reactors and must be considered in the process of selecting strategies and configurations for their fuel cycles [4, 5]. In addition to those requirements, proliferation resistance is among the core principles of nuclear power development and is an objective that should be fulfilled by advanced fuel cycles [6, 7]. Proliferation resistance also represents a necessary condition set by the European commission that needs to be met, in order the future advancement of nuclear energy in the European Union to be guaranteed [8]. It is thought that proliferation of nuclear materials is one of the most important problems standing before the future progress of nuclear energy [9]. Special attention to proliferation resistance has been paid since 2000 [10], the main concerns arising from plutonium stockpiles accumulation [11], and plutonium being considered among the most important materials related to non-proliferation [9].

Increased proliferation resistance would enhance nuclear power's growth outlook [12]. Currently, main factor in advanced fuel cycles' development is finding equilibrium between institutional measures (safeguards) and intrinsic material barriers [13]. Enhancing proliferation resistance would require implementation of internationally recognized and verified methodology for measuring proliferation resistance, as well as an integrated approach in applying simultaneously intrinsic barriers and safeguards. The need of such combined strategy is defined by the generally accepted view that simple technical solutions for improving proliferation resistance do not exist [12].

2 Nuclear Materials

Perhaps one of the most comprehensive definitions of nuclear materials is provided by the United States' Department of Energy (DOE). Their list includes: uranium isotopic mixtures with any amount of ^{235}U, ^{233}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, Bk, ^{252}Cf, Cm, enriched Li, ^{237}Np, Th, D, T, and all uranium located within enrichment cascades [14]. That list effectively includes almost all isotopes that can be used for rapid critical mass attainment. Below these isotopes are listed as in Ref. [15]: all plutonium isotopes, ^{235}U, ^{233}U, ^{237}Np, ^{231}Pa, ^{241}Am, ^{243}Am, ^{244}Cm, ^{245}Cm, ^{246}Cm, ^{247}Bk, and ^{251}Cf. The listed uranium isotopes could be used in a simplified "gun-type" weapon design, while ^{239}Pu usage requires that implosion technology be available. These technological specifications derive from the time needed to attain critical mass [15, 16]. The DOE also defines five classes of materials, according to their state, purity, and concentration of fissile isotopes: A – weapons materials, B – pure

materials (metal slabs, casts, *etc.*), C – high-purity materials (*e.g.* carbides, oxides, nitrides, alloys, mixtures), D – low-purity materials (*e.g.* low concentration solutions, residues, *etc.*), and E – all other materials [14].

3 Non-Proliferation and Physical Protection

Nuclear materials' non-proliferation is defined by the International Atomic Energy Agency (IAEA) as a property of a nuclear system or facility that impedes nuclear material's diversion and/or undeclared production, as well as malicious applications of nuclear technology by a state aiming to acquire nuclear weapon or other type of nuclear explosive device. That definition implies that proliferation resistance is a measure of the difficulties that should be overcome in order to develop and/or acquire nuclear weapon or nuclear explosive device by means of civilian fuel cycle and civilian technologies. Those difficulties include but are not limited to technical and technological barriers, required skills, time, *etc.* Misusing nuclear fuel cycle could include usage of materials, equipment, technological processes, facilities, and knowledge related to civilian fuel cycle [17]. In the case of nuclear proliferation the misuse of nuclear material and/or nuclear facilities is carried out by the owning country [18].

Sometimes, the meaning of 'proliferation resistance' is mistakenly associated with 'physical protection', the latter being such property of nuclear power system that hinders the theft of material suitable for constructing nuclear explosive device or device for spreading radioactive substances in the environment. Physical protection also impedes sabotages against nuclear facilities and prevents clandestine transportation of nuclear and radioactive material. In that case the perpetrator is a third party not owning the facilities and/or the material, be it non-national or national entity [17, 19]. Since the concept of 'material attractiveness' can be associated to both measures but in different context, it is essential that they be differentiated when proliferation resistance assessment is made [18–20].

4 Plutonium Classification and Usability

There are two main approaches for plutonium classification: the first relies on the ^{238}Pu concentration in the isotopic mixture since it defines the decay heat power of the plutonium ('*heat spike*'); the second classification is based on ^{240}Pu concentration. The decay heat power could be a limiting factor for the reliability and operability of a hypothetical nuclear explosive device, while ^{240}Pu defines mainly the spontaneous neutron emission: excessive amount of neutrons could initiate prematurely the chain reaction (the so called 'pre-detonation') that could lead to significant decrease of the yield of the hypothetical device ('*fizzle yield*') [21]. A classification based on ^{240}Pu concentration that includes five plutonium grades, and is commonly used, has been published by Pellaud [22] and can be seen in Table 1.

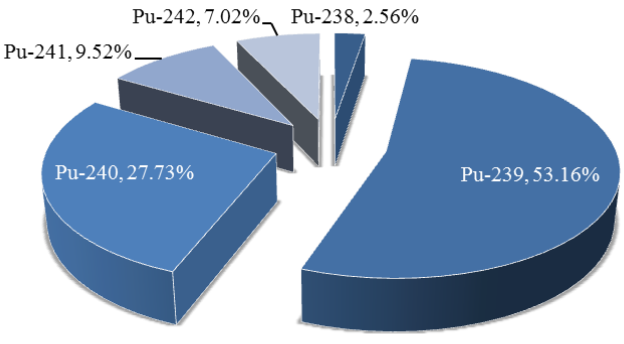

Figure 1. Isotopic vector of the plutonium, used for fresh MOX fuel fabrication [34].

Table 1. Plutonium grades, according to ^{240}Pu weight fraction [22]

Plutonium grades	**^{240}Pu fraction**	**Material's usability**
Super grade	$< 3\%$	Weapons material
Weapons grade	$3 - 7\%$	Weapons material
Fuel grade	$7 - 18\%$	Practically usable
Reactor grade	$18 - 30\%$	Possibly usable
MOX grade	$> 30\%$	Unusable

In order to define ^{238}Pu concentration threshold, above which the material is unusable for nuclear explosive device construction, impact assessments of increased decay heat power on material's usability and hypothetical device's reliability and operability have been carried out by Kessler et al. [25] and Kimura et al. [9]. As a result, the authors recommend minimal values of ^{238}Pu concentration in plutonium mixtures in order to render them unusable. Those values are summarised in Table 2.

Table 2. Minimal ^{238}Pu concentration that ensures the plutonium is unusable for weapon's manufacturing

	Kessler et al. [25]	**Kimura et al.** [9]
Low weapon technology	1.6 w%	2.0 w%
Medium weapon technology	3.2 w%	6.0 w%
High weapon technology	9.0 w%	15.0 w%

5 Proliferation Barriers

In order to define the level of civilian plutonium's usability for nuclear explosive device construction, it is necessary to analyse those properties of the material that could make it suitable for non-civilian applications. Most often, the attributes linked to material's proliferation resistance are the bare critical mass of a sphere, its decay heat, the spontaneous neutron emission, and the radiological barrier [3, 11]. Those factors compose the so called isotopic barrier to proliferation. Some of the factors influence material's usability, others – the probability for acquisition, transportation, and reprocessing [26]. The main properties of the most important plutonium isotopes are listed in Table 3.

The proliferation barriers can be divided in intrinsic and extrinsic [17, 26]. The intrinsic barriers may be physical – related to the properties of the material, and technical – related to the properties of the nuclear facilities, the access to the facilities, and the availability of specialists with

Table 3. Basic properties of main plutonium isotopes

Isotope	Half-life [22] [yrs.]	Bare critical mass [22] [kg]	Bare critical mass [21] [kg]	Bare critical mass [30] [kg]	Spontaneous neutron emission [22] [(n/kg)/s]	Decay heat power [22] [W/kg]	Dose rate* [34] [rad/h]
^{238}Pu	87.70	10.00	9.03	9.72	2,600,000	560.00	0.211000
^{239}Pu	24 100.00	10.00	10.23	10.11	22	1.90	0.000395
^{240}Pu	6 560.00	40.00	31.45	37.43	910,000	6.80	0.007170
^{241}Pu	14.40	10.00	12.24	13.09	49	4.20	0.001450
^{242}Pu	376 000.00	100.00	62.20	89.98	1,700,000	0.10	0.005450

*This is the dose rate of 20% of the critical mass at 1 m distance from the source.

adequate knowledge and skills, and time for device's development [17]. The most important factors of the intrinsic barrier are linked to material's properties; in total the isotopic, chemical, radiological, and weight factors have a relative share of 63.5 %. Other significant factor, with its relative weight of 18.1%, is the available inventory [26].

6 Objectives, Methodologies, and Input Data

6.1 Objectives

The main objectives of the current study are:

1. To evaluate the change in the material barrier of plutonium (but not its utility), obtained during normal operation of a reference commercial PWR, as a function of fuel type (UOX/MOX), burn-up (30,000 – 60,000 MWd/tHM), and cooling time (up to 30 years after discharge);
2. To try to compare different evaluation methods: isotopic criteria, 'Figure of Merit' methodology, and multi-attribute utility analysis.

6.2 Isotopic criteria

Since it is thought that aside of the countries, possessing nuclear weapons, only low weapons technology could hypothetically be present [25, 27], in the following assessment the low technology values of ^{238}Pu concentration, defined by Kessler et al. [25] and Kimura et al. [9] have been used as benchmarks (Table 2). Furthermore, the values of ^{240}Pu concentrations defining the reactor-grade and the MOX-grade plutonium (Table 1) have been used as an additional benchmark.

Although high spontaneous neutron emission results in higher pre-detonation probability, the values of ^{240}Pu concentration cannot be used as a standalone measure for the material barrier's quality for the following reasons:

- Pre-detonation precludes achieving nominal explosive yield of the device but does not eliminate a nuclear explosion [16, 28]: analyses carried out by Mark [23] and Kessler et al. [24, 25] show that if pre-detonation occurred in a hypothetical nuclear explosive device with nominal yield of 20 kt, the 'fizzle' yield would be between 0.12 kt and 0.54 kt (or between 0.6% and 2.7% of the nominal yield of the hypothetical device);
- Although ^{240}Pu is the most abundant even plutonium isotope in the examined vectors, it is not the only isotope contributing to plutonium's overall spontaneous neutron emission (see Table 3);
- Pre-detonation is a considerable factor that needs to be taken into account if the proliferator's goal is constructing a storable device with predictable yield [18] but is not by itself a limiting factor if the goal is to construct a device with any above-threshold yield (the threshold yield is the maximal explosive power achievable with conventional chemical explosives). It is accepted that pre-detonation even in low technology weapons would result in above threshold yields in most of the cases [29];
- Another factor is that even though the pre-detonation limits the explosive yield, the destruction radius is proportional to the cube root of the yield: a thousand-fold yield decrease leads to ten-fold decrease of the destruction radius [28]. That's why ^{240}Pu criteria had been selected only as an additional benchmark.

6.3 'Figure of Merit'

The 'Figure of Merit' methodology has been developed by a team from USA's Los Alamos National Laboratories. It is used to assess the material's usability for the construction of a nuclear explosive device by a nation or a sub-national group. The methodology uses two criteria, FOM_1 and FOM_2, in order to assess the material. They are calculated using Eqs. (1) and (2). These criteria include several intrinsic factors such as bare critical mass, spontaneous neutron fraction, decay heat, and dose rate. The first criterion assesses the material's usability when the explosive yield is of little importance and pre-detonation is not an issue, while the second criterion assesses the case when nominal yield and storability are desirable [18, 19, 29]. This effectively means that FOM_1 criterion does not take into account the effects of the spontaneous neutron emission, while FOM_2 could be used to assess the impact of even plutonium isotopes on the material barrier. The present analysis has been carried out using the algorithm applied in Ref. [30].

$$\mathrm{FOM}_1 = 1 - \lg\left[\frac{M}{800} + \frac{M \cdot DH}{4500} + \frac{M}{50}\left(\frac{DR}{500}\right)^{\frac{1}{\lg 2}}\right] \quad (1)$$

$$\mathrm{FOM}_2 = 1 - \lg\left[\frac{M}{800} + \frac{M \cdot DH}{4500} + \frac{M \cdot S}{6.8 \times 10^6} + \frac{M}{50}\left(\frac{DR}{500}\right)^{\frac{1}{\lg 2}}\right] \quad (2)$$

In Eqs. (1) and (2) M is the bare critical mass of a sphere in kg, DH is the specific decay heat in W/kg, S is the spontaneous neutron fraction in (n/s)/kg, and DR is the dose rate of 20% of the bare critical mass at a distance of 1 m from the source in rad/h. The higher the criteria value, the higher the material's usability [18, 19, 29]. The criteria values have approximate correspondence to DOE's material classes (Table 4). Some FOM reference values could be found in Refs. [18, 19, 29, 30]. In the present analysis the values of FOM_1 and FOM_2 are used as a metric to investigate the change in the material barrier's quality.

Table 4. Approximate correspondence between 'Figure of Merit' material evaluation and DOE classes [18]

FOM value	DOE class
> 2	$\sim$ B
[1;2]	$\sim$ C
[0;1]	$\sim$ D
< 0	$\sim$ E

6.4 Multi-attribute utility analysis (MAUA)

This tool has been developed by Charlton et al. as a means to assess the nuclear security (including both proliferation resistance and physical protection aspects) of nuclear processes. The analysis evaluates 14 utility functions, linked to proliferation barriers, each utility function having an assigned relative weight (Table 5). The utility functions, their assigned weights, and the complete calculation algorithm are fully described in Ref. [31]. The sum of the products of the relative weights and utility functions' values is used as a metric named '*static proliferation resistance*' and is calculated using Eq. (3); the sum value belongs to the interval [0;1]. The closer the value is to unity, the better is the quality of the barrier. This metric is applied in the present analysis in a similar fashion as the 'Figure of Merit' criteria – as a means to evaluate the change in the material barrier's quality.

$$PR_i = \sum_{j=1}^{J} w_j \, u_j(x_{ij}) \, . \quad (3)$$

$$NS = \frac{\sum_{i=1}^{I} m_i \, \Delta t_i \, PR_i}{\sum_{i=1}^{I} m_i \, \Delta t_i} \, . \quad (4)$$

In fact, the static proliferation resistance is used as a criterion to evaluate the proliferation resistance of the i-th stage of a nuclear process ($e.g.$ spent fuel reprocessing as a step in the closed fuel cycle). Then, using Eq. (4), the total nuclear security of the process can be evaluated. In the present analysis, the methodology is adapted, assuming that the nuclear process consists of a single stage: plutonium storage. Because the aim is to assess the material barrier, the storage time is disregarded, thus effectively equating the total nuclear security to the static proliferation resistance. Furthermore, in order to achieve relative comparability between the 'Figure of Merit' criteria and the static proliferation resistance, the value of some utility functions concerning physical protection aspects has been set to unity.

Table 5. Utility functions used in the multi-attribute utility analysis [31]

No	Utility function (u_i)	Unit of measure	Relative weight (w_{ij})
1	Attractiveness according to DOE (material class)	[—]	0.10
2	Material decay heat, due to plutonium	[W]	0.05
3	Relative weight fraction of even plutonium isotopes	[—]	0.06
4	Fissile material concentration	[SQ/t]	0.10
5	Dose rate	[(rem/h)/SQ]	0.08
6	Size/weight	[—]	0.06
7	Frequency of material's quantity measurement	[—]	0.09
8	Measurement uncertainty	[SQ/yr.]	0.10
9	Fissile material separability	[—]	0.03
10	Share of process steps, requiring material accounting	[—]	0.03
11	Probability for unidentified material movement	[—]	0.07
12	Physical barriers	[—]	0.10
13	Inventory	[SQ]	0.05
14	Type of fuel reloading	[—]	0.06

Unlike isotopic benchmarks and 'Figure of Merit' methodology that are applicable only to purified materials, $i.e.$ the assessment is carried out only for purified metallic plutonium, the multi-attribute utility analysis can be used to evaluate materials in various forms, including plutonium embedded in spent fuel ($i.e.$ not separated material) kept in spent fuel storage facility. That is due to the initial purpose of the multi-attribute analysis – to evaluate multi-stage nuclear processes where multiple material form transformations may occur. This allows evaluating and comparing the material barrier of purified metallic plutonium and non-separated metal that is contained within spent fuel assemblies.

In Eqs. (3) and (4) w_j is the relative weight of the j-th utility function, $u_j(x_{ij})$ is the value of the j-th utility function for the i-th stage of the nuclear process, PR_i is the static proliferation resistance of the i-th stage of the nuclear process, NS is the total nuclear security of the nuclear process, m_i is the mass of nuclear material in the i-th stage of the nuclear process, and Δt_i is the time period that the material spends in the i-th stage of the nuclear process.

6.5 Input data

The parameters of the reference PWR, used as input data in the analyses, are shown in Table 6. The isotopic compositions of the various plutonium mixtures and the material inventories have been calculated using the SCALE6.1

Table 6. Input parameters of the analysed reference PWR

Gross electric power	[MW]	1000
Gross thermodynamic efficiency of the power unit	[%]	32.6
Load factor of the power unit	[%]	85.0
Enrichment of uranium fuel	[%]	4.4
Plutonium weight share in MOX fuel	[%]	7.23

module ORIGEN-ARP by applying the standardised built-in cross-section library $w17\times17$ [32]. In order to calculate FOM_1 and FOM_2 values, the algorithm used in the analysis, presented in Ref. [30], has been applied, using the bare critical masses from Ref. [33].

Concerning the multi-attribute analysis, in order to neutralize the attributes, linked to physical protection and to differentiate between metallic plutonium and plutonium, embedded in spent fuel, the following values of several utility functions have been set:

1. For metallic plutonium:
   (a) DOE Attractiveness – $x_1 = 0$ (see Ref. [31]; metallic plutonium is material class B, according to DOE's classification [14]);
   (b) Size/weight – $x_6 = 0$;
   (c) Frequency of material's quantity measurement – $x_7 = 1$ (this function is not an aspect of the material barrier);
   (d) Fissile material separability – $x_9 = 0$ (the material is in metallic form, see Ref. [31]);
   (e) Share of process steps, requiring material accounting – $x_{10} = 1$ (this function is not an aspect of the material barrier; also, for the analysis' purposes it's been assumed that the examined nuclear process is a single-stage process);
   (f) Probability for unidentified material movement – $x_{11} = 1$ (recommended value by Charlton et al. [31]);
   (g) Physical barriers – $x_{12} = 0$ (it is assumed that the material is accessible because it is in metallic form; although the physical barriers are not aspects of the proliferation resistance, they are function of the material's form);
   (h) Type of fuel reloading – $x_{14} = 1$ (batch core loading is assumed, in spite of the material being in metallic form; that's because the frequency of obtaining plutonium depends on the core reloading pattern – batch or continuous. However, the same value for x_{14} has been set for embedded plutonium as well.);

Table 7. Main Plutonium isotopes' dose rates [36]

	X-ray dose rate [(mSv/h)kg]	Gamma dose rate [(mSv/h)kg]	Neutron dose rate [(mSv/h)kg]
^{238}Pu	5700.00	240.00	640.00
^{239}Pu	89.00	3.20	0.01
^{240}Pu	72.00	0.80	300.00
^{241}Pu	0.00	120.00	0.00
^{242}Pu	1.30	0.00	310.00

   (i) In order to calculate the value x_8 (utility function u_8 'Measurement uncertainty'), uncertainty of 0.2% is assumed, as recommended in Ref. [35]. For x_5 calculation (utility function u_5 'Dose rate') the values listed in Table 7 have been used.

2. For plutonium, embedded in spent fuel matrix:
   (a) DOE Attractiveness – $x_1 = 1$;
   (b) Size/weight – $x_6 = 1$;
   (c) Fissile material separability – $x_9 = 1$ (the material has not been separated from the spent fuel matrix);
   (d) Physical barriers – $x_{12} = 0.75$ (the material is in some type of spent fuel storage facility, see Ref. [31]);
   (e) Measurement uncertainty – $x_8 = 0$ (it is assumed that there is no uncertainty when item accounting is applied, $i.e.$ tracking down the number of items (fuel assemblies, spent fuel storage canisters, *etc.*) not the mass of the material itself, following the recommendation by Charlton et al. [31])

7 Results and Discussion

7.1 Isotopic criteria

The results of the benchmarking of the various examined plutonium vectors to different isotopic criteria are shown in Figures 2 – 5. The figures show the compliance of plutonium, obtained during UOX fuel operation with the criteria of Kessler et al. and Kimura et al. (Figure 2), the grade of plutonium, generated by UOX burning (Figure 3), the compliance of plutonium, obtained during MOX fuel operation with the criteria of Kessler et al. and Kimura et al. (Figure 4), and the grade of plutonium, generated by MOX burning (Figure 5).

Figure 2 unequivocally shows increased ^{238}Pu weight fraction with increased burn-up as well as its gradual decrease with cooling time. Those effects are expected, given the extended length of the fuel cycle, required for improving burn-up, and the subsequent decay of ^{238}Pu ($T_{½} = 84.4$ years). The important observations are linked to the impact those phenomena have on plutonium's material barrier. At the beginning of the cooling period – around the 110$^{\text{th}}$ day after discharge, the minimal burn-up at which the produced plutonium satisfies both criteria is 50,000 MWd/tHM. At the 30^{textth} year after fuel's discharge that is still the case although 50,000 MWd/tHM burn-up complies only marginally with the more stringent Kimura criterion. That clearly demonstrates the barrier deterioration over time. This effect, caused by ^{238}Pu weight fraction decrease, permits to conservatively assume that the minimal burn-up providing satisfactory compliance with both criteria at the 30$^{\text{th}}$ year following discharge is 55,000 MWd/tHM.

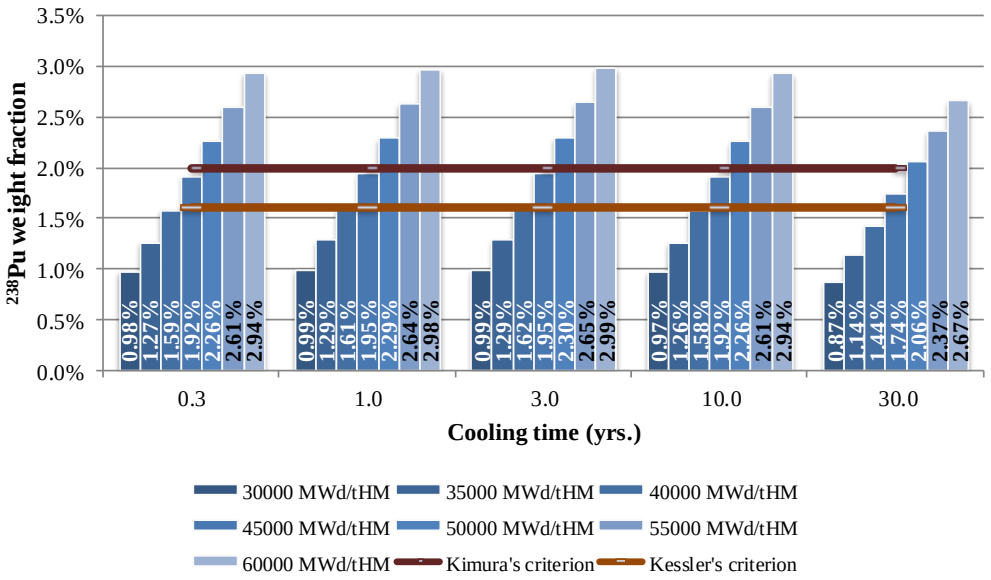

Figure 2. Correspondence of UOX fuel plutonium to Kessler and Kimura criteria.

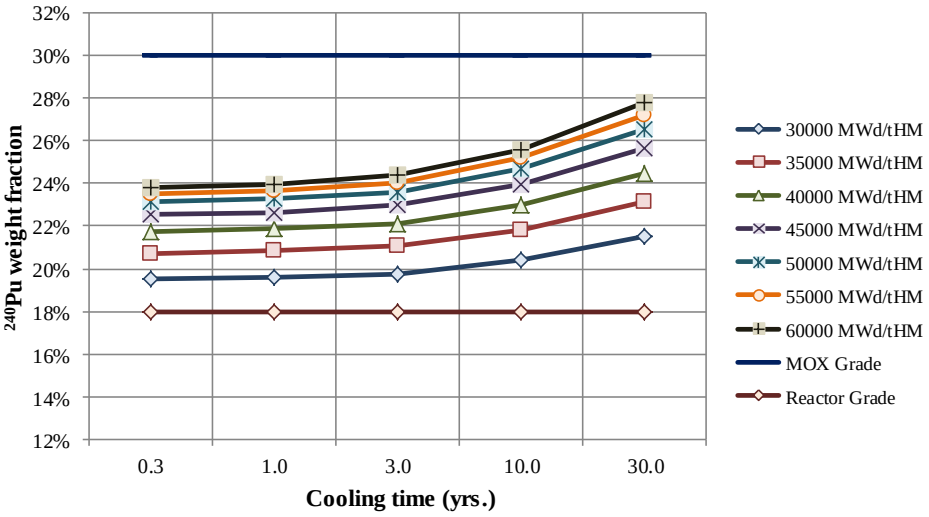

Figure 3. Correspondence of UOX fuel plutonium to plutonium grades.

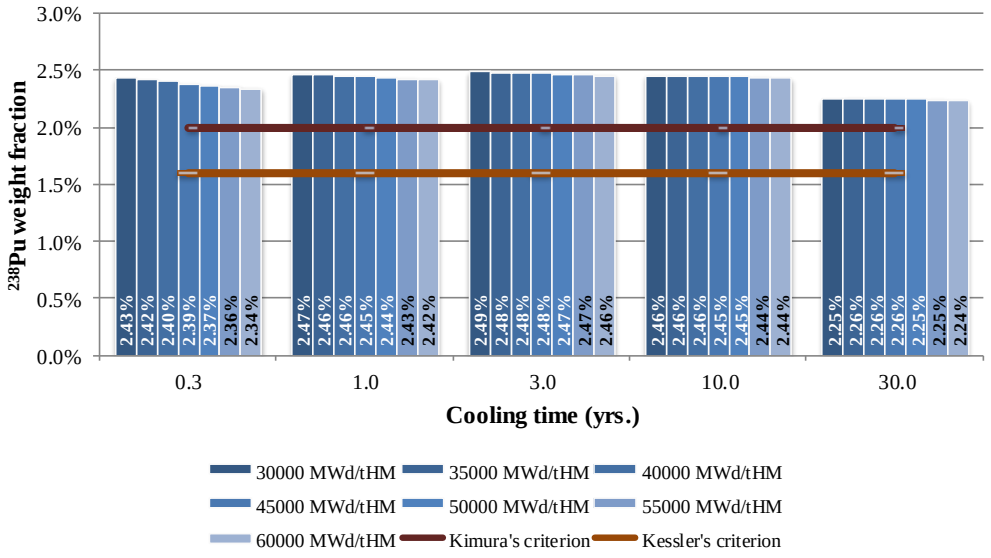

Figure 4. Correspondence of MOX fuel plutonium to Kessler and Kimura criteria.

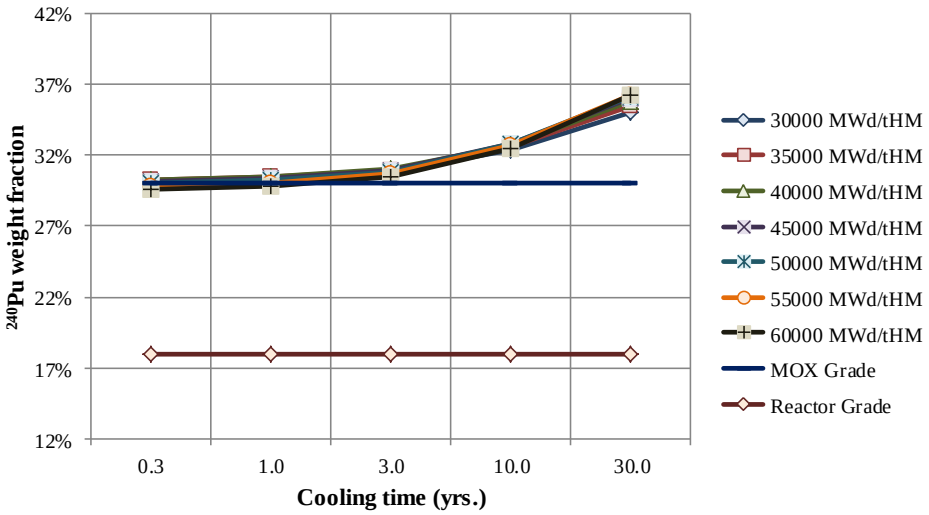

Figure 5. Correspondence of MOX fuel plutonium to plutonium grades.

Regarding the plutonium, obtained by MOX fuel irradiation, all examined mixtures have ^{238}Pu concentrations above the thresholds set by both criteria, irrespective of the burn-up and cooling time (Figure 4). Those results confirm the higher quality of MOX-grade plutonium's material barrier. That's due to the stability of ^{238}Pu concentration in the entire range of examined burn-ups and cooling time durations.

According to ^{240}Pu concentration, all plutonium mixtures, obtained from uranium fuel, fall into reactor-grade class (Figure 3). The observed trends for increasing ^{240}Pu concentration with burn-up and cooling time have been expected, since the concentrations of heavier plutonium isotopes in uranium fuel rise with irradiation, and the total plutonium mass diminishes with cooling time (see Ref. [33]), mainly due to ^{241}Pu decay ($T_{½} = 14.4$ years). The ^{240}Pu weight fraction increase leads to relative strengthening of the material barrier. However, as already noted in Section 6.2, this factor doesn't have independent influence. Regarding plutonium from spent MOX fuel, generally all investigated mixtures can be classified as MOX-grade (^{240}Pu weight fraction above 30%), with ^{240}Pu fraction increasing with cooling time. That phenomenon additionally improves MOX plutonium's material barrier quality, rendering the material virtually unusable for non-civilian usage.

Based on isotopic criteria benchmarking, the following conclusions can be formulated:

- The material barrier quality of plutonium from uranium spent fuel with low to medium burn-up (30,000 to 45,000 MWd/tHM) is not sufficient to independently guarantee the proliferation resistance of the material. However, that doesn't necessarily imply that utilizing this plutonium for non-civilian purposes might be feasible;

Table 8. Values of FOM_1 and FOM_2 criteria as a function of burn-up, cooling time, and fuel type

	Cooling time									
	FOM_1					FOM_2				
Burn-up [MWd/tHM]	0.3 [yrs.]	1.0 [yrs.]	3.0 [yrs.]	10.0 [yrs.]	30.0 [yrs.]	0.3 [yrs.]	1.0 [yrs.]	3.0 [yrs.]	10.0 [yrs.]	30.0 [yrs.]
	UOX									
30,000	2.39	2.39	2.39	2.40	2.42	1.29	1.28	1.28	1.27	1.26
35,000	2.34	2.33	2.33	2.34	2.35	1.22	1.22	1.21	1.20	1.18
40,000	2.27	2.27	2.27	2.28	2.30	1.16	1.16	1.15	1.14	1.12
45,000	2.23	2.22	2.22	2.22	2.24	1.11	1.11	1.1	1.09	1.07
50,000	2.17	2.17	2.17	2.17	2.19	1.06	1.06	1.05	1.04	1.02
55,000	2.12	2.12	2.12	2.12	2.13	1.02	1.01	1.01	0.99	0.96
60,000	2.08	2.08	2.08	2.08	2.10	0.97	0.97	0.97	0.95	0.93
	MOX									
30,000	2.09	2.09	2.08	2.09	2.09	0.90	0.90	0.89	0.87	0.84
35,000	2.09	2.09	2.08	2.08	2.09	0.89	0.89	0.88	0.86	0.82
40,000	2.08	2.08	2.07	2.07	2.08	0.87	0.87	0.86	0.84	0.80
45,000	2.08	2.08	2.07	2.07	2.08	0.86	0.85	0.85	0.83	0.78
50,000	2.08	2.08	2.07	2.07	2.07	0.85	0.84	0.84	0.81	0.77
55,000	2.08	2.07	2.06	2.06	2.06	0.84	0.83	0.82	0.79	0.74
60,000	2.08	2.07	2.06	2.06	2.06	0.83	0.82	0.81	0.78	0.73

- The easiest approach for improving the quality of the material barrier of plutonium from spent uranium fuel is increasing burn-up. That would add to the feasibility of increased burn-up fuels;

- The material barrier of plutonium from MOX fuel has significantly superior quality than that of plutonium from uranium fuel. However, the quality is weakly influenced by burn-up and cooling time.

7.2 'Figure of Merit'

The calculated FOM_1 and FOM_2 values are summarised in Table 8. The data show a general trend of decreasing criteria values, *i.e.* increasing quality of the material barrier. The values show relatively small barrier quality deterioration with cooling time. It is evident that burn-up has more pronounced influence on the quality of the barrier. The values of both criteria have been averaged over the 30-year spent fuel cooling period in order to analyse more thoroughly the effects of burn-up. The results are shown in Figure 6 and Figure 7. Figure 6 represents the change of the average FOM_1 value as a function of fuel burn-up. A clear, almost linear decrease in material's attractiveness can be observed for both plutonium from UOX and MOX spent fuels; in other terms, the material barrier's quality visibly improves with burn-up increase. The change in the quality of plutonium produced from uranium fuel is very well pronounced, especially when spontaneous neutron emission is not considered (FOM_1). The quality of the barrier of plutonium from spent MOX fuel basically remains constant with burn-up increase. Those results correspond with the trends outlined by the isotopic benchmarks (Section 7.1). The impact of the spontaneous neutron emission on the quality of the material barrier can be examined in Figure 7. In that case the quality of plutonium from spent MOX fuel also visibly increases with burn-up. Also, the FOM_2 values are around two times lower than FOM_1 values. That observation is unambiguous confirmation of the spontaneous neutron emission's contribution to material barrier's quality. However, the limited importance of the neutron emission (see Section 6.2) should be kept in mind.

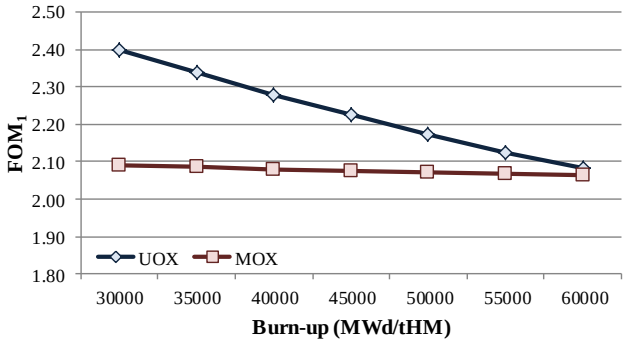

Figure 6. Change of 30-year average FOM_1 as a function of burn-up.

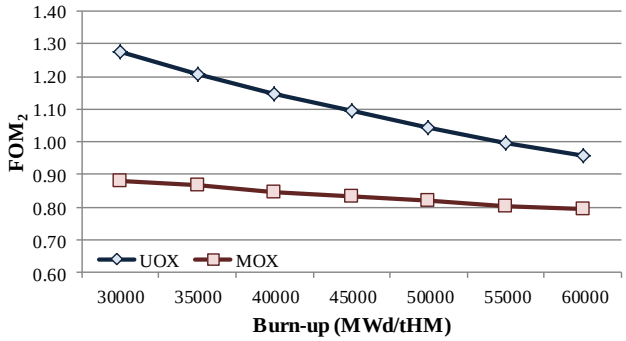

Figure 7. Change of 30-year average FOM_2 as a function of burn-up.

Table 9. Static proliferation resistance of metal plutonium and plutonium embedded in spent fuel matrix as a function of burn-up, cooling time, and fuel type

	Cooling time									
	Metal					Spent fuel				
Burn-up [MWd/tHM]	0.3 [yrs.]	1.0 [yrs.]	3.0 [yrs.]	10.0 [yrs.]	30.0 [yrs.]	0.3 [yrs.]	1.0 [yrs.]	3.0 [yrs.]	10.0 [yrs.]	30.0 [yrs.]
	UOX									
30,000	0.55	0.55	0.55	0.56	0.56	0.82	0.82	0.82	0.82	0.83
35,000	0.56	0.56	0.56	0.57	0.57	0.83	0.83	0.83	0.83	0.84
40,000	0.57	0.57	0.57	0.58	0.58	0.84	0.84	0.84	0.84	0.85
45,000	0.58	0.58	0.59	0.60	0.60	0.85	0.85	0.85	0.86	0.87
50,000	0.60	0.60	0.60	0.61	0.61	0.87	0.87	0.87	0.87	0.88
55,000	0.61	0.61	0.61	0.62	0.62	0.87	0.88	0.88	0.88	0.88
60,000	0.62	0.62	0.62	0.62	0.62	0.88	0.88	0.88	0.89	0.89
	MOX									
30,000	0.54	0.54	0.54	0.55	0.55	0.81	0.81	0.81	0.81	0.82
35,000	0.55	0.55	0.56	0.56	0.56	0.82	0.82	0.82	0.82	0.83
40,000	0.56	0.56	0.56	0.57	0.57	0.83	0.83	0.83	0.83	0.84
45,000	0.57	0.57	0.57	0.57	0.58	0.83	0.83	0.84	0.84	0.84
50,000	0.57	0.58	0.58	0.58	0.58	0.84	0.84	0.84	0.85	0.85
55,000	0.58	0.58	0.58	0.58	0.59	0.84	0.85	0.85	0.85	0.85
60,000	0.58	0.58	0.59	0.59	0.59	0.85	0.85	0.85	0.85	0.86

7.3 Multi-attribute analysis. Inventory and material form impact

The MAUA results for metallic and non-separated plutonium are shown in Table 9. Similarly to the 'Figure of Merit' values, the static proliferation resistance is virtually independent from cooling time. Because of that, similar approach is undertaken and the static proliferation resistance values are averaged over the 30-year cooling period. The dependence of static proliferation resistance from burn-up can be seen in Figure 8 for metallic plutonium and in Figure 9 for non-separated plutonium.

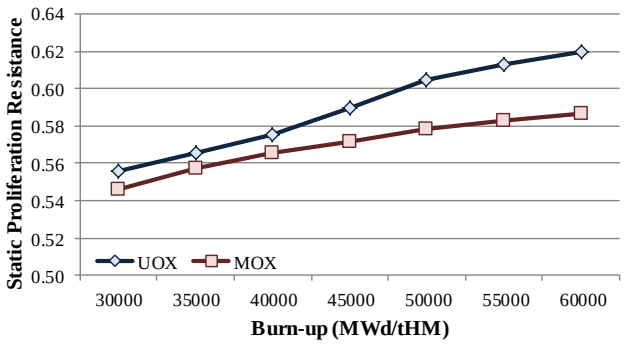

Figure 8. Change of 30-year average static proliferation resistance of separated plutonium as a function of burn-up.

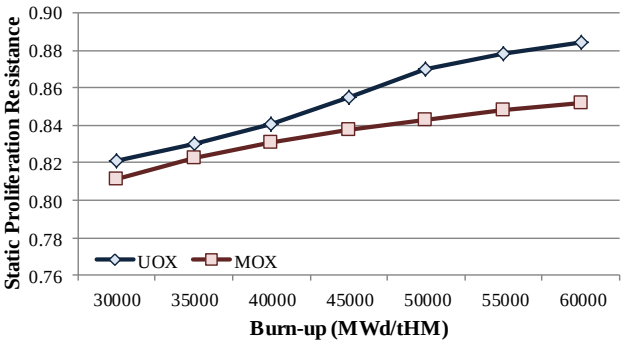

Figure 9. Change of 30-year average static proliferation resistance of non-separated plutonium as a function of burn-up.

The figures show that the character of the change of barrier's quality of metallic plutonium coincides with that of non-separated plutonium. Other significant observation is that, unlike the isotopic benchmarking and 'Figure of Merit' results, here the proliferation resistance of UOX plutonium is greater. The most likely reason for this discrepancy is the much greater plutonium inventory generated by mixed oxide fuel operation (Ref. [33]) which is considered in the MAUA analyses but is disregarded in isotopic benchmarking and 'Figure of Merit' assessments.

Other observation that can be made on the basis of the graphical data representation is that the improvement rate of MOX plutonium's barrier quality decelerates with burn-up increase. Concerning plutonium, generated from uranium fuel, the graph can be divided in two approximately linear sections with inflexion point at 50,000 MWd/tHM where the improvement rate decreases. When the material form is factored in, a higher rate of improvement with burn-up is observed for metallic plutonium (11.9% for plutonium from uranium fuel and 7.54% for plutonium from MOX fuel, in comparison with 7.7% and 4.9% for non-separated plutonium respectively). However, the contribution of the material form to the overall static proliferation resistance is significant and is illustrated in Figure 10. If the plutonium is embedded within the spent fuel matrix the relative quality of the barrier is between 42.8% and 48.5% higher depending on fuel type and burn-up. The relative contribution is greater for plutonium from spent mixed uranium-plutonium oxide fuel and is inversely proportional to the burn-up.

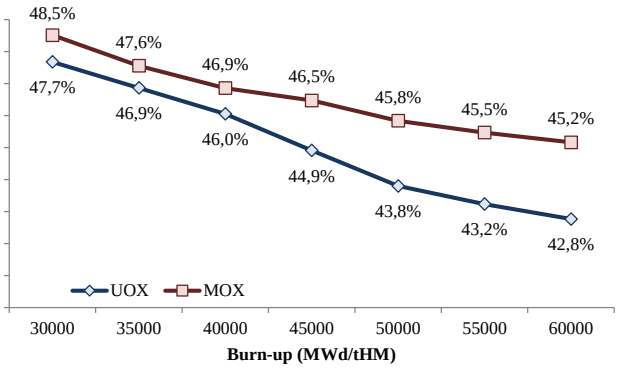

Figure 10. Relative increase of the static proliferation resistance of embedded plutonium compared to metallic plutonium.

8 Conclusions

An evaluation of the material barrier of plutonium generated in a reference PWR as a function of nuclear fuel's type and burn-up, and spent fuel's cooling time has been carried out using three different methodologies: isotopic benchmarking, 'Figure of Merit', and multi-attribute utility analysis. Based on the results the following conclusions can be formulated:

- Based on isotopic benchmarking the material barrier of plutonium obtained by SNF with low to medium burn-up (30–45 GWd/tHM) is not sufficient to render the material unattractive; however, that does not confirm plutonium's feasibility for non-civilian applications.
- The quality of the barrier of plutonium generated by UOX increases linearly with burn-up (faster isotopic vector degradation) which contributes to increased burn-up feasibility.
- If plutonium's inventory is not taken into account, the MOX plutonium material barrier quality is greater.
- Generally, burn-up has little impact on MOX plutonium barrier's quality.
- The impact of the considered cooling time period (up to 30 yrs.) in general is not significant which allows averaging the criteria values over the period.
- The protection by ^{240}Pu improves with cooling time but increased pre-detonation probability by itself

is not enough to substantially impact the barrier's quality since the potential yield of a hypothetical device would be above threshold levels.

- Given the results of the multi-attribute utility analysis, the material's inventory should not be disregarded in those assessments.
- The form of the material has unsurprisingly significant impact on the proliferation barrier – the plutonium embedded in SNF and stored within spent fuel assemblies/storage casks has nearly 50% better barrier quality.
- The rate of barrier quality increase with burn-up is higher for refined metal plutonium.
- Plutonium recycling in MOX improves overall barrier quality because material's decay heat, bare critical mass, dose rates, and neutron emission increase.
- Generally, the trends found by applying three different methods correspond well with each other (except the case when plutonium's inventory has been taken into account).
- Improved material barrier contributes to the proliferation resistance of nuclear fuel cycles. However, that doesn't imply weaker physical protection/safeguards.
- The physical protection analyses should take into consideration the properties of the material.
- The question of LWR reactor-grade plutonium's usability remains unresolved.
- The single recycling of plutonium, obtained by high-burn uranium fuel, in PWR in the form of MOX fuel with subsequent continuous storage of the secondary spent fuel appears to be an existing fuel cycle option that provides relatively good levels of material barrier quality.

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