

Plutonium-Containing Civilian Materials' Attractiveness Analysis Using the 'Figure of Merit' Methodology

I. Naydenov, K. Filipov

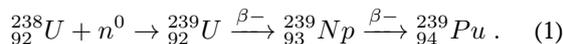
Technical University of Sofia, Department of Thermal and Nuclear Power Engineering, 8 Kliment Ohridski Blvd., 1000 Sofia, Bulgaria

Abstract. In the recent years, non-proliferation of nuclear materials has become an important issue, regarding civilian fuel cycle's development. The future nuclear energy systems' and nuclear fuel cycles' development should comply to non-proliferation criteria, and the materials that are used or discharged from the wide range of processes, involved in nuclear energy production, need to possess properties that make their usage in illicit nuclear explosives development extremely difficult, if not impossible. In the present article, a methodology, developed for assessing nuclear materials' attractiveness for diversion from the civilian fuel cycle, is applied. This is done in order to analyse the usability of some plutonium-containing materials from the civilian fuel cycle, most notably the plutonium that could be produced in commercial pressurised water reactors and subsequently extracted in reprocessing plants.

Keywords: figure of merit, nuclear fuel cycle, proliferation resistance, Plutonium

1 Plutonium in the Civilian Nuclear Fuel Cycle

Plutonium's presence in the civilian nuclear fuel cycle is due to its production in all types of nuclear reactors that utilize uranium-containing nuclear fuel. This is the result of a neutron capture by the ^{238}U nucleus, which after a series of beta decays, has as a product a ^{239}Pu nucleus, as it is shown in Eq. (1). Heavier plutonium isotopes are obtained by further neutron capture by plutonium nuclei.



Since the odd-numbered plutonium isotopes are fissile, the plutonium production in the power reactors is referred to as nuclear fuel breeding; that is the transformation of fertile isotopes into fissile ones. This process is described using the breeding ratio (BR). This ratio changes over the fuel campaign since some of the bred plutonium fissions. The breeding ratio depends on nuclear fuel's initial enrichment, reactor's physical properties, neutron energy spectra, etc., and is defined as a ratio between the amounts of newly produced fissile isotopes to the amount of initially loaded fissile material into the core.

The breeding ratio is an important characteristic of nuclear reactors. However, often for various economic and technical assessments, one needs to determine the total amount of plutonium that had been produced in a given reactor. This cannot be achieved solely by using the breeding ratio. That is why the plutonium build-up rate (or just plutonium rate) is used. That's the ratio between the amount of plutonium z and the amount of fission products α in the spent fuel:

$$PR = \frac{z}{\alpha} . \quad (2)$$

The amount of plutonium produced in a given reactor is proportional to the energy production, and using Eq. (3)

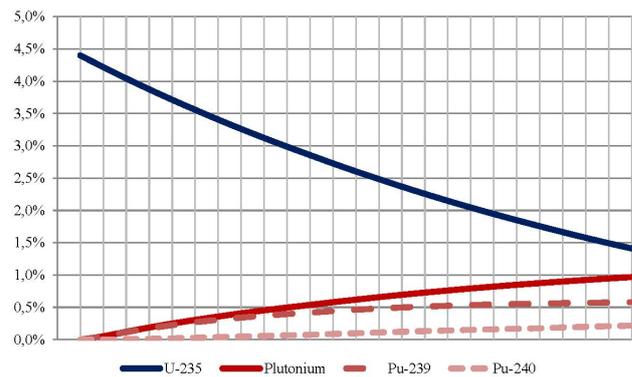


Figure 1. Uranium burn-up and plutonium build-up in pressurised water reactor.

the amount of plutonium could be easily calculated [1]. The plutonium build-up during the fuel campaign in pressurized water reactor is illustrated in Figure 1.

$$G_z = z G_x = \frac{PR \alpha W}{B \eta} . \quad (3)$$

The symbols used in Eq. (3) have the following meanings:

- G_z – total mass of plutonium, discharged annually with the spent fuel, [tHM];
- z – final plutonium mass concentration in the spent fuel;
- G_x – annual uranium loading, [tHM];
- α – final fission products mass concentration in the spent fuel;
- B – achieved burn-up, [MW•d/tHM];
- W – gross electricity production, [MW•h];
- η – unit's gross thermodynamic efficiency.

For that matter, there could be a significant amount of plutonium in power reactors' spent fuel. In fact, the annual plutonium generation amounts to between 136 and 366

kg per reactor, of which between 47 and 69% is ^{239}Pu , depending on the reactor type [2].

The plutonium's mass flow direction depends on the nuclear fuel cycle type. In once-through fuel cycle the discharged spent fuel is considered waste and eventually is to be disposed of in deep geological formations. In the case of closed or partially closed fuel cycle, all or part of the plutonium is recovered and reused in either light-water reactors or fast neutron reactors in a form, known as mixed uranium-plutonium oxide fuel (MOX).

2 Proliferation Resistance and Proliferation Paths

Proliferation resistance is a property of a nuclear material, nuclear facility or nuclear system that impedes the illicit production or diversion of nuclear material, as well as the purposeful usage of civilian facilities to produce weapons-grade nuclear material [3]. The materials that must be put under scrutiny are well defined by the U.S. Department of Energy and include all fissile uranium isotopes and mixtures, containing them, the plutonium isotopes from ^{238}Pu to ^{242}Pu , minor actinides, and thorium, among others [4]. Apart of the materials' composition, other very important feature vis-à-vis their international control, is their quantity. The IAEA defines the minimal controllable mass of plutonium as 8 kg, regardless its isotopic composition, unless the ^{238}Pu share is 80 w% or more. For the uranium these amounts are 8 kg for ^{233}U and 25 kg for ^{235}U . The U.S. NRC sets even lower values for the mass of nuclear materials – 5 kg for ^{235}U , and 2 kg for ^{233}U and ^{239}Pu [5].

Despite the presence of thorough international control (see Section 3), the possibility for civilian material and facilities misuse by a country or sub-national group cannot be dismissed. This could happen through theft, purchase of smuggled material and/or technology, material diversion, and breakaway from Nuclear Weapons' Non-proliferation Treaty [6]. The proliferation process is illustrated in Figure 2.

3 International Safeguards System and Nuclear Materials' Accountability

Since nuclear energy is considered dual purpose technology, it is necessary to apply some form of control over nuclear facilities and materials, although no nuclear-weapons state has produced its arsenal using civilian nuclear programme. However, there is an inherent proliferation risk that cannot be eliminated simply by restricting peaceful usage of nuclear energy [7]. Such a system is the IAEA's system of safeguards that provides control for materials and facilities alike. This system consists of three types of safeguards agreements between the Agency and the given country, as well as two types of protocols [8]. There exist other forms of international control as well such as the Nuclear Weapons' Non-proliferation Treaty that binds signatory countries not to acquire nuclear weapons and/or explosives if they are non-nuclear weapon country, to undertake steps towards nuclear disarmament, and to conclude an individual agreement with

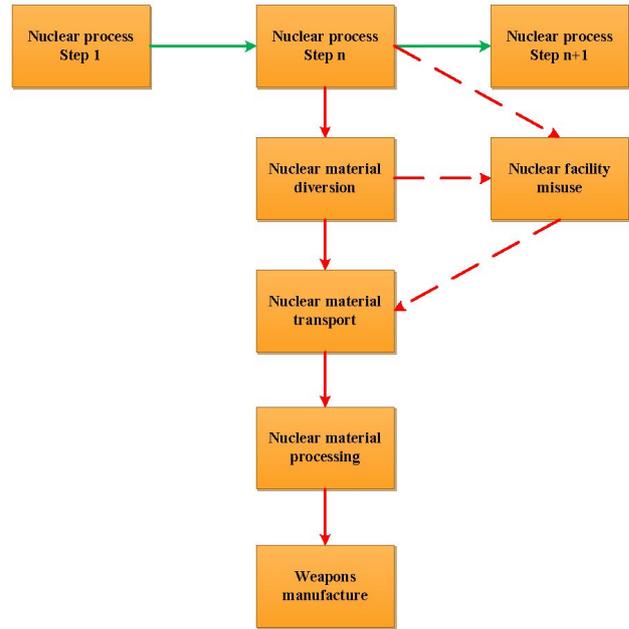


Figure 2. Nuclear material proliferation process [29].

the IAEA. An agreement between Euratom and the Agency is also in place. There is also an association of the states exporters of nuclear technology – the Nuclear Suppliers Group that enforces the nuclear technology recipient states to agree on special commitments that are based on non-proliferation criteria [7].

An important aspect of the safeguards system is the subjected nuclear material control. Therefore nuclear materials' accountability is one of the most important aspects of the international control and surveillance [9]. The IAEA has a comprehensive accountability system but other guidelines exist as well, *e.g.* the U.S. DOE standards [4].

4 Plutonium Properties

The physical and chemical properties of the nuclear materials that impede nuclear materials' proliferation and clandestine nuclear explosives' manufacturing are known as intrinsic barriers to proliferation [10]. The most important plutonium isotopes' properties are shown in Table 1.

Table 1. Plutonium properties, related to the intrinsic barrier

mass	Bare critical [11] [kg]	Decay heat [11] [W/kg]	SNF [12] [n s ⁻¹ kg ⁻¹]	Dose rate* [rad/h]
^{238}Pu	9.718	560.0	26 000	0.211000
^{239}Pu	10.110	1.9	22	0.000395
^{240}Pu	37.425	6.8	910 000	0.007170
^{241}Pu	13.086	4.2	49	0.001450
^{242}Pu	89.977	0.1	1 700 000	0.005450

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

There are two main ways to diminish the weapons quality of the material, thus its usability. The first approach for intrinsic barrier enhancement is to increase the amount of

decay heat, emitted by the isotopic mixture. This complicates the device using such a material, because it needs a cooling system. This also could affect the quality of the conventional explosive. This is achieved by increasing ^{238}Pu share. The second approach is based on increasing the spontaneous neutron fraction (SNF) that could lead to pre-detonation, thus diminishing the nuclear explosive's yield. This is done by augmenting the ^{240}Pu fraction.

5 Figure of Merit

The 'Figure of Merit' methodology has been developed by a team from USA's Los Alamos National Laboratories (LANL). 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 , to assess the materials' usability. They are calculated using Eqs. (4) and (5). 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. If for some reason the group, willing to construct the explosive device, desires higher yield, the yield of the device should be considered as well. In that case the second criterion is used [13,14]. The methodology assesses the material's usability for manufacturing a stable device that could be warehoused for some time. If the physical security of the materials is examined, then the first criterion should be applied, because it takes into account the factors that define the possibilities for acquiring, transporting, and processing the material. The methodology is used to assess metals, whether refined or not [12,15], although in this article non-irradiated plutonium dioxide is also considered.

The meaning of the criteria values is shown in Table 2 and typical FOM_1 values for the most common materials are listed in Table 3.

$$FOM_1 = 1 - \lg \left[\frac{M}{800} + \frac{M DH}{4500} + \frac{M}{50} \left(\frac{DR}{500} \right)^{\frac{1}{\lg 2}} \right] \quad (4)$$

$$FOM_2 = 1 - \lg \left[\frac{M}{800} + \frac{M DH}{4500} + \frac{M S}{6.8 \times 10^6} + \frac{M}{50} \left(\frac{DR}{500} \right)^{\frac{1}{\lg 2}} \right] \quad (5)$$

In Eqs. (4) and (5) M is the bare critical mass in kg, DH is the specific decay heat in W/kg, S is the spontaneous neutron fraction (SNF) in $n(\text{s kg})^{-1}$, 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 equations' terms have meanings as follows:

$$\begin{aligned} \frac{M}{800} & \quad \text{— size factor;} \\ \frac{M DH}{4500} & \quad \text{— stability factor;} \\ \frac{M S}{6.8 \times 10^6} & \quad \text{— yield factor;} \\ \frac{M}{50} \left(\frac{DR}{500} \right)^{\frac{1}{\lg 2}} & \quad \text{— acquisition factor.} \end{aligned}$$

Table 2. Meaning of FOM_1 and FOM_2 [12]

FOM	Material's Usability
> 2	Preferred material
1–2	Attractive material (precautions required)
0–1	Unpractical material (still possible use)
< 0	Unusable

Table 3. FOM_1 values for some nuclear materials [12]

Material	FOM_1	Material	FOM_1
^{233}U	2.70	^{242}Pu	1.90
^{235}U	2.20	^{242m}Am	2.60
^{236}Np	3.10	RG-Pu	2.13
^{237}Np	2.10	WG-Pu	2.73
^{238}Pu	0.90	$^{238}\text{Pu}/^{239}\text{Pu}$ (80:20)	1.01
^{239}Pu	2.80	20% enriched uranium	1.01
^{240}Pu	2.00	93% enriched uranium	2.18
^{241}Pu	2.60	^{233}U (^{232}U - 10 ppm)	2.69

6 Objectives

The future nuclear energy systems, in particular Generation IV systems, must implement proliferation resistant fuel cycles and their development should comply with non-proliferation criteria [18,19]. That means that the materials that are used or discharged from the wide range of processes, involved in nuclear energy production, need to possess properties that make their usage in illicit nuclear explosives development extremely difficult, if not impossible. Therefore, the objectives of the present paper are to assess different plutonium-containing civilian materials' theoretical usability for nuclear explosive manufacture. The analysis includes assessment of pure plutonium isotopes; plutonium compositions, obtained from PWR spent fuel in once-through and closed fuel cycle at different burn-ups; plutonium dioxide, containing different grades of plutonium. Other objective is analysis of spent fuel plutonium's usability as a function of burn-up.

It is very important to underline that this analysis assesses the potential quality of already purified material in metallic form and of plutonium dioxide, containing several types of plutonium mixtures. The isotopic compositions are calculated by using models for reference of WWER and PWR reactor types. The assessment is purely theoretical and aims to prioritise civilian materials with regards with their physical security and the need for safeguards application. The data used as input is not derived from any operational power reactor; instead we have used averaged typical values for fuel enrichment, load factor, thermodynamic efficiency, burn-up, etc. The two models are used for comparison purposes only, they are only numerical models, and do not refer to any real reactor, power plant, or country, neither have other implications whatsoever.

It should also be kept in mind that the examined materials are assumed to have been already purified and they are not in the form they are discharged, *i.e.* irradiated fuel assemblies that are being stored in spent fuel pools or in wet or dry storage facilities.

7 Input Data

In order to analyse different mixtures and plutonium-containing compounds, it is needed to know their isotopic compositions, bare critical masses, and other properties, related to the intrinsic proliferation barrier.

7.1 Isotopic compositions' calculation

The isotopic compositions of plutonium in spent fuel as a function of burn-up are calculated using the module ORIGEN-ARP, which is part of the software code SCALE, developed by Oak Ridge National Laboratories (ORNL) [16]. The reactors data, used for the calculations, are presented in Table 4. In Table 4 N represents the power unit's installed gross capacity, φ is unit's load factor, η is unit's gross thermodynamic efficiency, y is the tails assay from enrichment, c_5 is the enrichment of uranium fuel and c_{Pu} is the plutonium's share in the MOX fuel. The results are outlined in Table 5, Table 6 and Table 7. The fuel burn-up

Table 4. Reactor properties

	N [MW]	φ [%]	η [%]	y [%]	c_5 [%]	c_{Pu} [%]
PWR	1000	85	32.6	0.3	4.4	7.23
WWER	1000	85	32.6	0.3	4.4	—

is represented in MWd/tHM. The fuel burn-up ranges from 30 000 MWd/tHM to 65 000 MWd/tHM in order to allow analysis of spent fuel, produced in different generations of nuclear reactors that achieve different burn-ups. Moreover, this range allows the analysis of isotopic composition's variation as a function of burn-up and helps assessing materials' usability dependence on burn-up increase. This would help designing proliferation resistant fuel cycles that would be beneficial in other aspects as well, *e.g.* increased energy production and more efficient natural resources exploitation, and will comply with Generation IV technological requirements.

In the present article only pressurised water reactors' fuel cycle has been reviewed because these are the most common types of nuclear reactors around the world. They represent 63% of all nuclear reactors and 68.3% of the gross installed capacity [17].

The initial plutonium fraction in the fresh MOX fuel is 7.23% reactor-grade plutonium. It is assumed that the MOX fuel is manufactured from plutonium, obtained from LWR spent fuel that achieved 45 000 MWd/t burn-up and has been kept in interim storage for cooling for 10 years (the exact composition is shown under Table 8 and is sourced from [12]), and from depleted uranium with 0.3% ^{235}U content.

Table 5. Isotopic composition of plutonium that could be extracted from spent WWER UOX fuel right after discharge as a function of burn-up

	30 000 [MWd/tHM]	35 000 [MWd/tHM]	40 000 [MWd/tHM]	45 000 [MWd/tHM]	50 000 [MWd/tHM]	55 000 [MWd/tHM]	60 000 [MWd/tHM]	65 000 [MWd/tHM]
^{238}Pu	0.955%	1.254%	1.583%	1.938%	2.312%	2.693%	3.072%	3.437%
^{239}Pu	62.746%	59.093%	55.938%	53.202%	50.827%	48.763%	46.953%	45.376%
^{240}Pu	18.636%	19.505%	20.192%	20.739%	21.175%	21.518%	21.802%	22.034%
^{241}Pu	14.306%	15.659%	16.599%	17.199%	17.526%	17.644%	17.617%	17.474%
^{242}Pu	3.357%	4.489%	5.689%	6.921%	8.160%	9.380%	10.557%	11.679%

Table 6. Isotopic composition of plutonium that could be extracted from spent PWR UOX fuel right after discharge as a function of burn-up

	30 000 [MWd/tHM]	35 000 [MWd/tHM]	40 000 [MWd/tHM]	45 000 [MWd/tHM]	50 000 [MWd/tHM]	55 000 [MWd/tHM]	60 000 [MWd/tHM]	65 000 [MWd/tHM]
^{238}Pu	0.994%	1.298%	1.634%	1.994%	2.370%	2.755%	3.136%	3.505%
^{239}Pu	63.058%	59.460%	56.337%	53.626%	51.271%	49.218%	47.423%	45.849%
^{240}Pu	18.333%	19.230%	19.953%	20.546%	21.030%	21.423%	21.746%	22.025%
^{241}Pu	14.338%	15.660%	16.593%	17.195%	17.535%	17.675%	17.663%	17.547%
^{242}Pu	3.277%	4.352%	5.482%	6.639%	7.795%	8.928%	10.032%	11.075%

Table 7. Isotopic composition of plutonium that could be extracted from spent PWR MOX fuel right after discharge as a function of burn-up

	30 000 [MWd/tHM]	35 000 [MWd/tHM]	40 000 [MWd/tHM]	45 000 [MWd/tHM]	50 000 [MWd/tHM]	55 000 [MWd/tHM]	60 000 [MWd/tHM]	65 000 [MWd/tHM]
^{238}Pu	2.405%	2.380%	2.358%	2.336%	2.313%	2.292%	2.270%	2.249%
^{239}Pu	41.280%	39.878%	38.633%	37.570%	36.656%	35.902%	35.295%	34.794%
^{240}Pu	30.096%	30.127%	30.079%	29.947%	29.754%	29.498%	29.189%	28.868%
^{241}Pu	16.668%	17.446%	18.097%	18.603%	18.996%	19.255%	19.405%	19.447%
^{242}Pu	9.551%	10.169%	10.833%	11.545%	12.281%	13.053%	13.842%	14.642%

Table 8. Plutonium isotopes' bare critical masses

	[3]	[21]	[22]	[23] [‡]	[24]	[12] [#]	[11]	[25] [†]	[26]		calculated
	[kg]	[kg]	[kg]	[kg]	[kg]	[kg]	[kg]	[kg]	min [kg]	max [kg]	
²³⁸ Pu	10.000	9.700	10.000	9.600	7.100	9.700	9.030	13.100	8.946	9.490	9.718
²³⁹ Pu	10.200	10.100	10.000	10.160	10.000	10.000	10.230	14.800	9.990	10.331	10.110
²⁴⁰ Pu	36.800	36.900	37.000	36.540	33.000	37.300	31.450	44.800	35.700	39.033	37.425
²⁴¹ Pu	12.900	13.000	13.000	12.670	12.400	13.000	12.240	17.600	12.270	13.042	13.086
²⁴² Pu	89.000	83.400	89.000	88.760	85.000	89.100	62.200	87.800	85.600	85.600	89.977
WG-Pu	—	10.700	—	10.620	—	10.500	—	—	—	—	—
RG-Pu	—	—	—	13.480	—	14.400	—	—	—	—	—

[‡] In Ref. 23 the exact isotopic compositions of both weapons-grade and reactor-grade plutonium are not clarified.

[#] Plutonium density – 19.84 g/cm³; weapons-grade plutonium composition: ²³⁸Pu – 0.01%; ²³⁹Pu – 94.026%; ²⁴⁰Pu – 5.814%; ²⁴¹Pu – 0.13%; ²⁴²Pu – 0.02%; reactor-grade plutonium composition at 45 000 MWd/t burn-up and after 10 years decay: ²³⁸Pu – 2.56%; ²³⁹Pu – 53.16%; ²⁴⁰Pu – 27.73%; ²⁴¹Pu – 9.52%; ²⁴²Pu – 7.02%.

[†] δ -phase, density – 15.8 g/cm³

From the results listed in Table 5 and Table 6 becomes evident that with increasing burn-up ²³⁹Pu share decreases and all other isotopes' shares increase. Regarding spent MOX fuel (Table 7), similar trends can be observed. However, there are few differences – ²³⁸Pu share slightly diminishes, and the share of ²⁴⁰Pu decreases as well.

7.2 Critical masses' calculation

The critical masses of all materials are calculated for spherical geometry, assuming homogenous mixture with zero porosity, using the computer software KENO V.a. [18]. Fresh plutonium dioxide's critical masses are calculated assuming that the compound is stoichiometric.

Pure plutonium isotopes' calculated bare critical masses are listed in Table 8, alongside values from different sources. It is easy to conclude that the calculated values correspond very well with the values obtained from literature sources.

The results from critical masses calculation for fresh PuO₂ with different plutonium isotopic composition and for different plutonium mixtures, produced in power reactors, are shown in Table 10 and Table 11. The calculations for PuO₂ are carried out for three types of the compound – containing only ²³⁹Pu, containing weapons-grade plutonium, and containing reactor-grade plutonium. Weapons-grade and reactor-grade plutonium compositions are given under Table 8. The densities used in the calculations are 10 970 kg/m³ for PuO₂ [27] and 19 840 kg/m³ for other plutonium mixtures [12]. The PuO₂ composition is shown in Table 9.

Table 9. PuO₂ composition

	²³⁹ PuO ₂ [w%]	RG-PuO ₂ [w%]	WG-PuO ₂ [w%]
²³⁸ Pu	0.0000	2.2429	0.0088
²³⁹ Pu	88.1919	46.7701	82.9046
²⁴⁰ Pu	0.0000	24.4989	5.1478
²⁴¹ Pu	0.0000	8.4458	0.1156
²⁴² Pu	0.0000	6.2626	0.0179
Oxygen	11.8081	11.7797	11.8055

Table 10. Different PuO₂ bare critical masses

²³⁹ PuO ₂ [kg]	WG-PuO ₂ [kg]	RG-PuO ₂ [kg]
28.351	29.480	40.318

Table 11. Bare critical masses of plutonium mixtures that could be extracted from PWR spent nuclear fuels as a function of burn-up

		WWER [kg]	PWR [kg]	PWR MOX [kg]
30 000	[MWd/tHM]	12.941	12.726	15.799
35 000	[MWd/tHM]	13.177	13.086	16.014
40 000	[MWd/tHM]	13.453	13.490	16.131
45 000	[MWd/tHM]	14.054	13.714	16.282
50 000	[MWd/tHM]	14.108	13.827	16.574
55 000	[MWd/tHM]	14.439	14.284	16.809
60 000	[MWd/tHM]	14.595	14.673	16.638
65 000	[MWd/tHM]	14.989	14.752	16.895

As a result, it is evident that with decreasing ²³⁹Pu share, the critical mass increases, mainly due to increased ²⁴⁰Pu and ²⁴²Pu share. At higher burn-ups these isotopes are between a third and a quarter of the total plutonium.

7.3 Other Plutonium properties

The other properties are calculated using Eqs. (6), (7), and (8), where c_i is the weight share of the i -th plutonium isotope [28].

$$DH = \sum c_i \cdot DH_i \quad (6)$$

$$S = \sum c_i \cdot S_i \quad (7)$$

$$DR = \sum c_i \cdot DR_i \quad (8)$$

8 Results and Discussion

The results of the usability criteria calculation are outlined in Table 12, Table 13 and Table 14.

Table 12. FOM₁ and FOM₂ values for pure plutonium isotopes

	FOM ₁		FOM ₂
	calculated	Ref. 9	calculated
²³⁸ Pu	0.9131	0.9	0.9001
²³⁹ Pu	2.7720	2.8	2.7711
²⁴⁰ Pu	1.9858	2.0	0.2914
²⁴¹ Pu	2.5441	2.6	2.5426
²⁴² Pu	1.9413	1.9	-0.3543

The calculated values for FOM₁ for pure plutonium isotopes agree very well with the values obtained by Bathke [12]. As a result, the fissile isotopes of plutonium are assessed as preferred materials, ²⁴⁰Pu and ²⁴²Pu are attractive materials, and ²³⁸Pu is unpractical material but still its use is possible (see Table 2). Considering high-yield explosives, ²⁴²Pu is unusable material (the only material with negative value for any of the criteria) because of its very high number of spontaneous neutrons (Table 1). ²⁴⁰Pu is unpractical material, although the low FOM₂ value suggests its usage is unlikely. Considering other isotopes, the fissile isotopes have unsurprisingly high usability, and ²³⁸Pu requires scrutiny in spite of the fact it is in the category of "unpractical materials".

Regarding the fresh PuO₂ (Table 13), all of the considered compositions are usable with usability increasing with ²³⁹Pu share increase. That's true for all plutonium-containing materials. Only the PuO₂ that contains reactor-grade plutonium from point of view of creating high-yield explosive. The first two PuO₂ (containing pure ²³⁹Pu and weapons-grade plutonium) compositions have been assessed in order to obtain reference values.

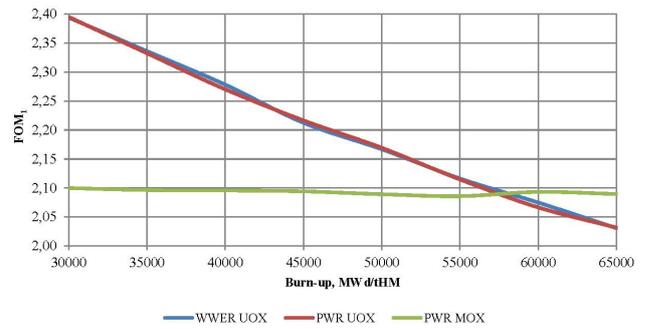
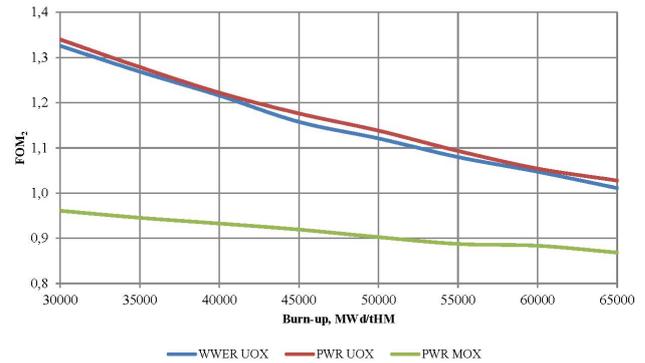
 Table 13. FOM₁ and FOM₂ values for different grades of PuO₂

	FOM ₁	FOM ₂
²³⁹ PuO ₂	2.3373	2.3365
WG-PuO ₂	2.5026	1.5946
RG-PuO ₂	1.7234	0.6684

The main points of the analysis of the usability of plutonium, produced in pressurized water reactors are two: (1) assessing the relationship between usability and achieved burn-up, and (2) analysing the influence of the type of fresh fuel on material's usability.

 Table 14. FOM₁ and FOM₂ values for plutonium mixtures that could be extracted from PWR spent nuclear fuels as a function of burn-up

		WWER UOX		PWR UOX		PWR MOX	
		FOM ₁	FOM ₂	FOM ₁	FOM ₂	FOM ₁	FOM ₂
30 000	[MWd/tHM]	2.3940	1.3258	2.3950	1.3398	2.0999	0.9608
35 000	[MWd/tHM]	2.3359	1.2687	2.3324	1.2787	2.0964	0.9452
40 000	[MWd/tHM]	2.2782	1.2158	2.2702	1.2221	2.0957	0.9326
45 000	[MWd/tHM]	2.2125	1.1576	2.2164	1.1762	2.0941	0.9192
50 000	[MWd/tHM]	2.1669	1.1209	2.1692	1.1381	2.0892	0.9025
55 000	[MWd/tHM]	2.1162	1.0796	2.1146	1.0933	2.0859	0.8875
60 000	[MWd/tHM]	2.0748	1.0472	2.0664	1.0542	2.0932	0.8835
65 000	[MWd/tHM]	2.0306	1.0111	2.0315	1.0277	2.0895	0.8684
AVERAGE	—	2.2011	1.1534	2.1995	1.1663	2.0930	0.9125


 Figure 3. FOM₁ of plutonium mixtures, extracted from different types of spent fuel, as a function of burn-up.

 Figure 4. FOM₂ of plutonium mixtures, extracted from different types of spent fuel, as a function of burn-up.

The FOM₁ and FOM₂ values for plutonium, produced in pressurized water reactors, are shown in Table 14, and their change as a function of burn-up is illustrated in Figure 3 and Figure 4. The usability of plutonium from spent uranium fuel sharply drops with burn-up increase. Still, for the considered burn-up range, all of the plutonium has FOM₁ greater than 2, which makes it preferred material. Clearly, spent uranium fuel should be strictly scrutinized, and this currently is being done. On average, FOM₁ is around 2.2 for both WWER and other PWR types of reactors, with WWER's plutonium value slightly higher. This clearly shows that plutonium, produced in commercial PWRs is clearly usable in nuclear explosive device, although the usability decreases with burn-up increase. As a whole, the average FOM₁ values for both WWER and PWR plutonium from spent UOX fuel are slightly higher than the value for reactor-grade plutonium, obtained in [12] (see Table 3). That can be explained by the fact that the



Figure 5. Plutonium-containing materials order according to their usability, assessed with FOM₁ criterion.



Figure 6. Plutonium-containing materials order according to their usability, assessed with FOM₂ criterion.

plutonium's compositions considered in the present article are those at the moment of discharge from the core, whereas the value in Table 3 is for reactor-grade plutonium that has been cooled down for 10 years.

Considering FOM₂, where the hypothetical explosive's yield is also a factor, it also decreases with burn-up increase. In this case, the values approach 1, which makes the material attractive and requiring control (Figure 4).

Regarding the closed fuel cycle application, using MOX fuel, the plutonium obtained at the end of the fuel campaign falls into the "preferred material" category all along the burn-up range. Maybe more surprisingly, the FOM₁ has little to no change with burn-up increase. At very high burn-ups this material is more attractive than plutonium, produced from uranium fuel. The average FOM₁ value is almost 2.1, which is slightly less than the respective value for UOX fuel. All plutonium, obtained from spent MOX fuel, however, is impractical for usage in high-yield devices, and the FOM₂ gradually decreases below 0.9 for burn-ups over 50 000 MWd/tHM.

Based on the calculated FOM₁ and FOM₂ values for different materials, the examined materials are arranged in descending order in terms of their usability. In Figure 5 the arrangement is according to FOM₁ criterion. The plutonium, produced in commercial PWRs comes right after the pure fissile isotopes of plutonium and PuO₂, containing pure ²³⁹Pu and weapons-grade plutonium. The only material that is in neither "preferred" nor "attractive" material is pure ²³⁸Pu. All other examined materials are at least "at-

tractive", seven of them being "preferred" with commercially produced plutonium firmly in this category.

FOM₂ arrangement is almost identical (Figure 6) to that, according to FOM₁. Three of the considered materials are in the "preferred" category and all of them contain only fissile material, two being pure fissile isotopes. The reactor-grade plutonium containing PuO₂ and pure ²⁴⁰Pu are impractical, and ²⁴²Pu is unusable for high-yield explosive device. All other materials, including those produced in commercial reactors, are "attractive" materials that should be under control.

9 Conclusion

As a conclusion, we could state that the plutonium, produced in commercial pressurised water reactors is attractive material for use in some kind of nuclear explosive and the increase in burn-up and the fuel cycle closure decrease but do not eliminate this attractiveness. However, the 'Figure of Merit' methodology assesses the materials' usability only as a function of some components of the intrinsic proliferation barriers and the fact that this plutonium should be extracted from the spent fuel assemblies, transported, and processed in form, suitable for nuclear explosive manufacture, which is no easy task, should be taken into account.

It is necessary to underline that the successful application of the international safeguards system that's currently in place must be continued in spite of the increasing burn-ups and nuclear cycle closure that decrease the material's quality for nuclear explosives production.

Acknowledgments

The research presented in the current paper has been supported by Technical University of Sofia's research subsidy (PhD Student Support Contract № 152ПД0005-02).

References

- [1] Velev V., Filipov K., Nuclear Fuels, IFO Design, Sofia (2008) (in Bulgarian).
- [2] IDAHO NATIONAL LABORATORIES, Plutonium Discharge Rates and Spent Nuclear Fuel Inventory Estimates for Nuclear Reactors Worldwide (2012).
- [3] INTERNATIONAL ATOMIC ENERGY AGENCY, Technical Features to Enhance Proliferation Resistance of Nuclear Energy Systems, Nuclear Energy Series, No. NF-T-4.5, IAEA, Vienna (2010).
- [4] U.S. DEPARTMENT OF ENERGY, Nuclear Material Control and Accountability, Manual DOE M 470.4-6 (2006).
- [5] Artisyuk V., Saito M., Ezoubtchenko A., Development of methodology to assess proliferation resistance of nuclear heavy metals, *Progress in Nuclear Energy* 50 (2008) 647-653.
- [6] AMERICAN PHYSICAL SOCIETY, Nuclear Power and Proliferation Resistance: Securing Benefits, Limiting Risk, A report by the Nuclear Energy Study Group of the American Physical Society Panel on Public Affairs (May 2005).
- [7] Kessler G., Proliferation-Proof Uranium/Plutonium Fuel Cycles. Safeguards and Non-proliferation, KIT Scientific Publishing, Karlsruhe (2011).

- [8] INTERNATIONAL ATOMIC ENERGY AGENCY, IAEA Safeguards Serving Nuclear Non-Proliferation, Vienna (2015).
- [9] INTERNATIONAL ATOMIC ENERGY AGENCY, Nuclear Material Accounting Handbook, IAEA-SVS-15, Vienna (2008).
- [10] Skutnik S. E., Yim M.-S., Assessment of fuel cycle proliferation resistance dynamics using coupled isotopic characterization, *Nuclear Engineering and Design* **241** (2011) 3270-3282.
- [11] Permana S., Suzuki M., Basic evaluation on material attractiveness of isotopic plutonium barrier, *Progress in Nuclear Energy* **53** (2011) 958-963.
- [12] Bathke C.G., Material Attractiveness and Why It Is Important, Los Alamos National Laboratory, LA-UR-14-20797 (February 2014).
- [13] King W.E. *et al.*, The application of a Figure of merit for nuclear explosive utility as a metric for material attractiveness in a nuclear material theft scenario, *Nuclear Engineering and Design* **240** (2010) 3699-3707.
- [14] Bathke C.G. *et al.*, An Assessment of the Attractiveness of Material Associated with a MOX Fuel Cycle from a Safeguards Perspective, In: *Proceedings of INMM 50th Annual Meeting*, Los Alamos National Laboratory (2009).
- [15] Bathke C.G., Further Assessments of the Attractiveness of Materials in Advanced Nuclear Fuel Cycles from a Safeguards Perspective, Los Alamos National Laboratory, LA-UR-08-05958, 10th Information and Exchange Meeting "Actinide and Fission Product Partitioning and Transmutation", Mito, Japan, October 6-10 (2008).
- [16] OAK RIDGE NATIONAL LABORATORIES, SCALE: A Comprehensive Modeling and Simulation Suite for Nuclear Safety Analysis and Design, ORNL/TM-2005/39, Version 6.1 (June 2011).
- [17] Naydenov I., Filipov K., LWR Fuel Cycles' Material and Isotopic Balance, *BgNS Transactions* **20** (2015) 62-69.
- [18] U.S. DEPARTMENT OF ENERGY, A Technology Roadmap for Generation IV Nuclear Energy Systems, GIF-002-00 (2002).
- [19] OECD NUCLEAR ENERGY AGENCY, Technology Roadmap Update for Generation IV Nuclear Energy Systems (2014).
- [20] OAK RIDGE NATIONAL LABORATORIES, KENO V.a: An Improved Monte Carlo Criticality Program, ORNL/TM-2005/39, Version 6.1, Sect. F11 (2011).
- [21] Kang J., von Hippel F., Limited Proliferation Resistance Benefits from Recycling Unseparated Transuranics and Lanthanides from Light-Water Reactor Spent Fuel, *Science and Global Security* **13** (2005) 169-181.
- [22] U. S. NATIONAL ACADEMY OF SCIENCES, Monitoring Nuclear Weapons and Nuclear-Explosive Materials: An Assessment of Methods and Capabilities, Appendix A (2005).
- [23] Magill J., Peerani P., (Non-) Proliferation Aspects of Accelerator Driven Systems, *J. Phys. IV France* **9** (1999) 167-181.
- [24] Clayton E.D., Anomalies of Nuclear Reactivity, Revision 6, Pacific Northwest National Laboratory, Richland, WA, PNNL – 19176 (February 2010)
- [25] Kimura, Y. *et al.*, Evaluation of proliferation resistance of plutonium based on spontaneous fission neutron emission rate, *Annals of Nuclear Energy* **46** (2012) 152-159.
- [26] INSTITUT DE RADIOPROTECTION ET DE SÛRETÉ NUCLÉAIRE, Evaluation of nuclear criticality safety data and limits for actinides in transport, Final Report, C4/TMR2001/200-1 (2013).
- [27] OAK RIDGE NATIONAL LABORATORIES, Thermophysical Properties of MOX and UO₂ Fuels Including the Effects of Irradiation, Fissile Materials Disposition Program, ORNL/TM-2000/351 (2000).
- [28] Permana S. *et al.*, Analysis on isotopic plutonium barrier based on spent nuclear fuel of LWR, *Annals of Nuclear Energy* **75** (2015) 116-122.
- [29] Cleary V.D. *et al.*, Strengthening the Foundations of Proliferation Assessment Tools, Sandia National Laboratories, SAND2007-6158 (September 2007).