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A new scientific solution for preventing the misuse of reactor-grade plutonium as nuclear explosive

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The authors dedicate this paper to Prof. Dr. Wolf Häfele, the initiator of safeguards research at Karlsruhe, who followed their research into non-proliferation with particular interest.

ABSTRACT

Yield analyses of nuclear explosions and thermal analyses of hypothetical nuclear explosive devices (HNEDs) based on reactor-grade plutonium are examined in a common approach. Three different levels of HNED technology are defined by criteria of geometric dimensions and thermodynamic characteristics of the chemical high-explosive implosion lenses. The results show the content of Pu-238 and the heat it generates in reactor-grade plutonium to be the key parameter. Low-technology HNEDs based on reactor-grade plutonium from spent low-enriched uranium (LEU) or MOX LWR fuel with burnups of 30 GWd/t or more are technically unfeasible. For medium technology, this limit rises to approximately 55 GWd/t burnup. Special cooling applied to such HNEDs would increase these burnup limits still further. Higher Pu-238 contents in reactor-grade plutonium are required to make such HNEDs technically unfeasible. Only for high-technology HNEDs, which could only be built by Nuclear Weapon States (NWSs), the limit to the Pu-238 content of reactor-grade plutonium would rise to approximately 9%.

The paper discusses scientific lower limits of alpha-particle heat power or Pu-238 contents above which reactor-grade plutonium can be considered denatured or proliferation-resistant. However, eventually such limits could only be determined by IAEA in agreement with the countries concerned.

Such denatured, proliferation-resistant reactor-grade plutonium, which makes reactor-grade plutonium HNEDs technically unfeasible, can be produced by various fuel cycle strategies employing enriched reprocessed uranium (ERU) or minor actinides (MAs). An interim phase of denatured proliferation-resistant plutonium production can be envisioned. A fully proliferation-resistant civil plutonium fuel cycle will become possible later. The use of MAs creates additional proliferation problems. While americium cannot be misused for weapon purposes, neptunium may well be. The neptunium actinide, therefore, must be avoided in an appropriate strategy of a future proliferation-resistant civil nuclear fuel cycle. A fuel cycle strategy of this type is proposed.

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1. Introduction

After the International Fuel Cycle Evaluation Program (INFCE, 1980), the USA decided around 1980–1982 to give up reprocessing of spent LWR fuel and refabrication of plutonium/uranium mixed oxide (MOX) fuel as well as the use of MOX fuel in nuclear reactors. It had become apparent by then that the amount of plutonium produced in LWRs in the USA and elsewhere by 1980 exceeded the amount of weapon plutonium in the nuclear weapon states (NWSs). At the same time, leading US scientific organizations and US authors, e.g., the National Academy of Sciences (1995) or Garwin (1998) stated that reactor-grade plutonium could be used

for nuclear weapons. These statements were repeated frequently. They are summarized in Fig. 1:

Also the fast reactor (FR) programs were given up in several countries in the following decade. Declared US national nuclear waste policy was direct spent fuel disposal. Germany adopted the same waste management policy for its nuclear fuel cycle in 2005. Other countries, such as France or Japan, did not follow this line.

More recent discussions in the USA envisage retrievability of conditioned spent fuel from geologic disposal sites to provide for a later turnaround of fuel cycle policy (GNEP, 2006). Also, new research programs including recycling strategies for the nuclear fuel cycle (use of reprocessing and fuel refabrication) were initiated (Laidler, 2003; US Department of Energy, 2008).

A number of publications, e.g., by DeVolpi (1979), Pellaud (2002) and DeVolpi et al. (2004) either expressed doubts about the validity of the statement quoted above or complained that it was not scientifically sound, let alone justified (Dautray, 2001). In Kessler et al. (in press), the authors attempted to find a scientific basis for these

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Plutonium of the quality produced in current nuclear power reactors, following separation from spent fuel and purification, can be used to make nuclear explosives, using technology comparable to that of the earliest plutonium weapons. While weapons made from this plutonium would have much less reliable and considerably lower explosive yields than similar weapons made from weapons-grade plutonium, they remain nevertheless highly dangerous nuclear explosives.

Fig. 1. Statement about misuses of reactor-grade plutonium.

statements and the issues it entailed. They analyzed the thermodynamic characteristics of so-called hypothetical nuclear explosive devices (HNEDs) based on reactor-grade plutonium. There was found to be a limit for reactor-grade plutonium containing a certain percentage of Pu-238 at which such devices would become technically unfeasible (Kessler, 2007). The reason is that the heat produced by Pu-238 would cause the chemical high explosives around the fissile material either to melt or to ignite spontaneously in a chemical reaction. Denatured reactor-grade plutonium of this type could be produced in various fuel cycle options investigated in (Broeders and Kessler, 2007). They require closed fuel cycles with reprocessing and refabrication of the fuel. All plutonium can be burnt except for unavoidable losses in chemical reprocessing and refabrication, which must be disposed of together with the fission products (Kessler, 2002). These new fuel cycle strategies would allow a proliferation-resistant denatured reactor-grade plutonium fuel cycle and present plutonium, now mainly kept in interim storage, to be burnt (Broeders and Kessler, 2007). Even the minor actinides could be burnt provided the necessary chemical separation processes were applied on a technical scale (Herbig et al., 1993; Laidler et al., 1997; Conocar et al., 2006). The minor actinides could also be taken care of by recycling in different reactors, e.g., LWRs, fast-spectrum reactors (FRs), or accelerator driven systems (ADSs) (Kessler, 2002). However, this raises questions of possible misuse of the neptunium and americium. Kessler (2008), therefore, analyzed the proliferation resistance of americium originating from various fuel cycle options. Americium, which is always generated in spent reactor fuel as a mixture of Am-241, Am-242m, and Am-243, was found to be proliferation-resistant. The case of neptunium, which occurs almost exclusively as Np-237, is more difficult. It cannot be denatured by other isotopes and it has practically no alpha-particle heat power. It can only be avoided by combining the thorium fuel cycle and the plutonium fuel cycle (Kessler et al., 2007). The main findings of the analyses will be outlined below.

Analysis of the potential nuclear explosive energy yield of reactor-grade plutonium

Kessler et al. (in press) used the similarity between early safety analysis and disassembly theory for uranium and plutonium fast neutron criticals as well as early experimental fast reactors as described by Bethe et al. (1956), McCarthy et al. (1958), Stratton et al. (1958), Hummel et al. (1960), and Nicholson (1964). While the theories of nuclear excursion, reactivity feedbacks, and the description of disassembly or explosion are directly applicable to nuclear explosions of plutonium-based HNEDs, the input data and power excursion characteristics differ drastically:

- The input values for the reactivity ramps caused by implosioninduced shock compression are several orders of magnitude higher in an HNED than in safety analysis of nuclear reactors.
- There is no Doppler coefficient in HNEDs because the neutron energy spectrum is so hard as to be above the resonance region.

- No coolant or fuel expansion feedback reactivities occur in HNEDs for the microseconds of a nuclear explosion.
- During power excursion of a nuclear explosion power densities rise to such levels as to reach internal pressures in the [TPa] range.

2.1. Verification of theoretical models and equation of state

The authors verified their theoretical models and materials characteristics by recalculating the example of a nuclear explosion published only in part (Sandmeier et al., 1972). The authors' results were close to the published results. The authors also confirmed Serber's formula for calculating nuclear explosion yields for various maximum excess reactivities achieved by shock compression during implosion (Serber, 1992). On this basis, the authors went on calculating the potential nuclear explosion yield of reactor-grade plutonium by using the validated models and materials data.

2.2. Differences between nuclear explosive devices and HNEDs based on reactor-grade phitonium

The nuclear excursion in nuclear explosive devices is ignited by an external neutron source immediately upon complete shock compression of the fissile material. Consequently, ignition of the power excursion must occur at maximum supercriticality of the device.

An HNED based on reactor-grade plutonium has a strong inherent spontaneous fission neutron source multiplied according to the subcriticality of the device. This leads to early pre-ignition of the power excursion immediately upon prompt criticality. Power rises fast very soon leading to internal energies and pressures high enough to stop the inward progress to the center of the HNED of the shock wave caused by the high-explosive lenses. The shock wave therefore compresses only the outer parts of the fissile sphere. This limits excess reactivity and, finally, the potential nuclear explosion yield.

The only analysis ever published of a calculated estimate of the potential nuclear explosive yield of HNEDs (Mark, 1993) with reactor-grade plutonium (from reprocessed spent fuel with a burnup of 30 GWd/t) was not based on a detailed numerical analysis, but most probably used inside knowledge from Los Alamos and the Serber formula (Serber, 1992). It assumed pre-ignition at the earliest possible point when shock compression leads to prompt criticality, and it applied the criterion of the shock wave being stopped when the aggregate internal fission energy released in power excursion is equal to the energy generated by the surrounding chemical explosives. Application of Serber's formula then furnishes a minimum fizzle nuclear explosive yield of 0.54 kt TNT (equivalent) (Mark, 1993).

2.3. Authors' calculations of the nuclear explosive yield of HNEDs

The following Pu-vector was taken as an example: 5.5% Pu-238, 34.1% Pu-239, 31.1% Pu-240, 10.6% Pu-241 and 18.7% Pu-242 (ρ = 15.8 g/cm³ for the δ -phase of plutonium). This composition had been named Pu-(1) by (Kessler, 2007). Taking $k_{\rm eff}$ = 0.98, calculations yield a radius of 5.8 cm for a solid plutonium sphere encased by a 5-cm reflector of natural uranium (see Fig. 2). The weight of this subcritical sphere is approximately 13 kg. Neutron lifetime is $\ell_{\rm eff}$ = 1.4 × 10 8 s for this uncompressed fissile material arrangement. If the plutonium sphere is compressed to twice its original density, neutron lifetime decreases to $\ell_{\rm eff}$ = 1.3 × 10 $^{-8}$ s.

Shock compression of the plutonium sphere and natural uranium reflector system were treated by the theoretical tools also applied to inertial confinement fusion (Duderstadt et al., 1982). This resulted in density changes within the HNED as a function

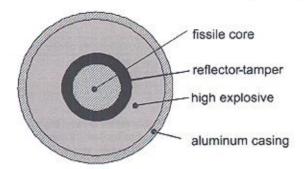


Fig. 2. Geometric arrangement and dimensions for scoping studies of critical masses for reactor-grade plutonium.

of space and time for different pressures acting on the outside reflector surface. Additional parametric Monte Carlo calculations finally produced the Rossi alpha $\alpha(t)$ for the shock wave progressing inward. The pre-ignition theories of Mark (1993) and Seifritz (2007) show that the relatively high inherent total fission neutron source leads to early pre-ignition roughly 1 μs after prompt criticality. The probability density function for the differential probability of pre-ignition is very narrow, thus making the nuclear energy yields quasi-deterministic (Seifritz, 2007).

Table 1 shows the results of our calculations of the nuclear explosive yield for two cases of 0.06 TPa (low technology) and 0.11 TPa (very high technology) produced by the high explosive lenses and acting on the outer surface of the natural uranium reflector. Plutonium boiling would start at 11 μs in the 0.06 TPa and at 8.35 μs in the 0.11 TPa cases. In the 0.06 TPa case, the shock wave would be stopped at 11.47 μs at 4-cm radius of the plutonium sphere when a Rossi alpha of $\alpha(t)$ = 9.26 μs^{-1} would be reached. In the 0.11-TPa case, the shock wave would be stopped at 9.307 μs at 4.5-cm radius of the plutonium sphere when a Rossi alpha of $\alpha(t)$ = 12.72 μs^{-1} would be attained.

The maximum explosive energy yield of approximately 0.12 kt TNT (equivalent) is reached at approximate 12.7 μ s in case of 0.06 TPa. In case of 0.11 TPa, a maximum explosive energy yield of around 0.35 kt TNT (equivalent) is attained approximately at 10.7 μ s.

Discussion of our results and comparison with those of Mark (1993)

Our results of 0.12 kt TNT (equivalent) for the 0.06 TPa and 0.35 kt TNT (equivalent) for the 0.11 TPa cases quantitatively are below the 0.54 kt TNT (equivalent) published by (Mark, 1993). The differences can be explained to some extent by Mark's cruder criterion of the internal fission energy released being equal to the energy of the high explosive lenses, and by the use of Serber's formula. Our results differ from those of Mark in quantitative though not in qualitative terms. These results based on perfect spherical symmetry of the shock wave acting on the outer surface of the reflector exceed the explosive yield of all known conventional chemical explosive devices. Besides the explosive yields also radiation and radiotoxicity effects of such nuclear explosions must be taken into account.

Table 1
Maximum nuclear explosive yield for the low technology and very high technology

Case	Max. nucl. expl. yield (kt) TNT (equivalent)	Reached at time (µs)
0.06 TPa (low technology)	0.12	12.7
0.11 TPa (very high technology)	0.35	10.7

These findings result from neutronic investigations. Next, the results of thermal analysis will be discussed together with the question whether such HNEDs are technically feasible under the geometric dimensions and levels of technology assumed.

3. Thermal analysis of HNEDs at different levels of technology

In the previous chapter we had selected the Pu(1) reactor-grade plutonium composition from Kessler (2007) because it represents an upper limit in determining the nuclear explosive yield for UOX LWR spent fuel up to a burnup of $60\,\mathrm{GWd/t}$ or MOX fuel recycled (up to three times with a burnup of $50\,\mathrm{GWd/t}$). This plutonium composition leads to the highest mass (k_{eff} =0.98) in an HNED and, therefore, to the highest potential nuclear explosive yields (Kessler, 2007) compared to plutonium compositions originating from reprocessing spent UOX fuel with a burnup below $60\,\mathrm{GWd/t}$ or MOX fuel with a burnup below $50\,\mathrm{GWd/t}$.

In this chapter, the results of neutronic analyses are shown to not necessarily imply that such HNEDs are technically feasible. This point will be proved by thermal analysis. After some minor correction to the alpha-particle heat power of Pu-242 (Chebeskov, 2007) the heat produced by HNEDs will be used as the main leading parameter. This alpha-particle heat power is associated with a specific composition of reactor-grade plutonium, in which the Pu-238 isotope dominates the heat output. Moreover, various levels of technology required to design HNEDs will be defined and discussed in the light of the neutronic and thermal analyses conducted above.

3.1. Definition of different levels of technology

The geometric dimensions of the reactor-grade plutonium sphere and its reflector are determined by the nuclear characteristics of the fissile core and the reflectors. They vary between 5.3 and 5.8-cm radius for a solid reactor-grade plutonium sphere and $k_{\rm eff}$ = 0.98 when reflected by a 5-cm thick natural-uranium reflector shell (Kessler, 2007). This applies to the reactor-grade plutonium compositions arising from different fuel cycle options (Broeders and Kessler, 2007). Hollow reactor-grade plutonium spheres would have an outer diameter of approximately 7.2 cm.

Different levels of technology are required to design a nuclear explosive device with reactor-grade plutonium, e.g., fabrication of the plutonium metal to a specific purity with respect to light elements, like boron, carbon, oxygen, or design and construction of the implosion lenses with different high explosives, etc. Only the different levels of technologies needed for neutronic and thermal analyses will be discussed here.

3.1.1. Level of technology required for neutronic analysis

In Section 2.3, two levels of technology and the associated pressures were defined:

- low technology 0.06 TPa;
- · very high technology 0.11 TPa.

These levels of technology incorporate the art of designing and building the high explosive lenses and are directly related to the thickness of the explosive lenses. The increase in pressure applied to the outside of the reflector is a function of convergent spherical flow and has been shown (Kessler et al., in press) to be the higher the thicker the high explosive lenses are. The higher the magnification factor or the pressure applied to the outer surface of the reflector, the higher are the particle velocities caused by the shock wave, the maximum Rossi alpha attained and, finally, also the nuclear explosive yields.

Table 2
Geometric dimensions and thermodynamic data of explosion lenses

Level of technology	Thickness of implosion lenses (cm)	Thermal conductivity (W/m°C)	Melting temperature (°C)	Temperature (°C) for starting self-explosion
Low technology	43	0.219-0.494	79	180-288
Medium technology	25	0.259-0.452	190-286	260-322
Very high technology	10	0.561	448	331-347

3.1.2. Level of technology required for thermal analysis

Thermal analysis in Kessler (2007) showed the temperature increase in the high explosive lenses to be higher, with more disadvantageous effects, for thicker high explosive lenses. This result points in the direction opposite to that desired in nuclear explosive yield analysis (Section 3.1.1 above).

Accounts of the development of nuclear weapons in Rhodes (1995), Cochran et al. (1987), and Podwig (2004) indicate the outside radius of nuclear weapons to have decreased from 65 cm (earliest nuclear weapon) to some 22 cm (Fetter et al., 1990). At the same time, the thickness of high explosive lenses decreased from 43 cm (Rhodes, 1995) to approximately 25 cm to some 10 cm (Fetter et al., 1990). The three levels of technology assumed in Kessler (2007) (see Table 2) are based on this development.

Table 2 and Kessler (2007) show the levels of technology to be based not on only geometric dimensions but also on the thermodynamic data of the materials used in HNEDs, e.g., the high explosive lenses. These thermodynamic data are much more important in reactor-grade plutonium HNEDs than in weapons-grade plutonium nuclear explosive devices. The heat produced by Pu-238 is negligible (less than 20 W) in a weapons-grade plutonium based nuclear explosive device, thus requiring no thermal analysis. For reactor-grade plutonium HNEDs, however, these thermodynamic data are decisive, as will be seen below. The most critical parts are the high explosive lenses.

As the geometric shape of the high explosive lenses is unknown, a so-called one-dimensional conservative approach was chosen in which the real temperature profile in the explosive lenses always tends to be underestimated (Kessler, 2007).

The history of nuclear weapons development (Cochran et al., 1987; Rhodes, 1995; Podwig, 2004) shows that a non-nuclear weapon state (NNWS) about to design and build HNEDs from reactor-grade plutonium would have to proceed from low to medium technology levels. Only current nuclear weapon states (NWSs) would be able to master the very high level of technology because of their know-how in research, experiments, and development accumulated over long periods of time.

The levels of technology applicable to thermal analysis will be discussed below.

3.2. Low technology

A low-technology HNED with low-technology dimensions (65 cm outer radius and 43 cm radial thickness of the high explosive lenses), high explosives of low technology (Tables 2 and 3), and reactor-grade plutonium was investigated in Kessler (2007). According to Kessler et al. (in press) this would allow a concentric pressure shock of approximately 0.18 TPa to act on the outer

surface of the reflector and probably result in even higher nuclear explosive yields than those reported in Section 2.3 above. However, this HNED would not be technically feasible, as is borne out by the results of thermal analysis of this HNED.

The results of thermal analysis (Fig. 3 and Table 2) are based on a subcritical radius of 5.3 cm for $k_{\rm eff}$ = 0.98 for reactor-grade plutonium with approximately 2.1% Pu-238 and a total alpha-particle heat power of 0.144 kW. Baratol and Composition B were selected as high explosives for the earliest nuclear weapons (Rhodes, 1995). Fig. 3 shows that some 20–40% of the volume of high explosives would melt and lose its geometric structure. Calculations show that even with an alpha-particle heat power around 0.120 kW corresponding to Pu-(0) (Kessler, 2007), with a plutonium composition of 1.6% Pu-238, 58.8% Pu-239, 20.8% Pu-240, 13.8% Pu-241, 5% Pu-242, from reprocessing of spent UOX LWR fuel with a burnup of 30 GWd/t, some 10–20% of the high explosive lenses would melt.

Consequently, all reactor-grade plutonium arising from reprocessed spent LWR UOX or MOX fuel with burnups slightly below 30 GWd/t to more than 60 GWd/t, or all reactor-grade plutonium generating more than 0.120 kW in an HNED would render a lowtechnology HNED technically unfeasible. The nuclear explosive yields of 0.12 or 0.35 kt TNT (equivalent) in Section 2.3 would not be feasible either.

This result does not confirm the statement in Fig. 1 which reads:

...plutonium of the quality produced in current nuclear reactors...can be used to make nuclear explosives, using technology comparable to the earliest plutonium weapons.

The first nuclear weapon had exactly the geometric dimensions, and contained the chemical explosives, (Rhodes, 1995) shown in Fig. 3 and Table 3 above. However, it incorporated a weapons-grade plutonium sphere with a slightly smaller radius (approximately 4.8 cm instead of 5.3 cm, see Fig. 3). It did work with weapons-grade plutonium, but it would not work with reactor-grade plutonium producing more than 0.120 kW in its fissile part.

As described in Cochran et al. (1987) and Podwig (2004), the development of nuclear weapons very quickly proceeded to more efficient and smaller nuclear explosive devices. Therefore, mediumtechnology HNEDs will be examined next.

3.3. Medium technology

A medium-technology HNED based on the assumptions of Kessler (2007) is shown in Fig. 4. It has an outer radius of 42 cm and high explosive lenses 25 cm in thickness, which would produce a shock pressure of 0.11 TPa (case B in Kessler et al., in press) on the outer surface of the reflector. This would imply an explosive yield of approximately 0.35 kt TNT (equivalent) (Section 2.3).

 $\begin{tabular}{ll} Table 3 \\ Characteristic materials data for low technology high explosives \\ \end{tabular}$

High explosive	Density (g/cm ³)	Thermal conductivity (W/m°C)	Detonation velocity (km/s)	Melting point (°C)	Onset of pyrolysis (°C)	Temperature for onset of chemical explosion $T_{\rm in}$ (°C)	Thermal decomposition energy (kJ/kg)
Baratol	2.61	0.494	4.9	79-80	180	300	1256
Composition B	1.74	0.219	7.8-8.0	79	200	214	1256-2094
TNT	1.45	0.259	6.9	81	260	288	1256

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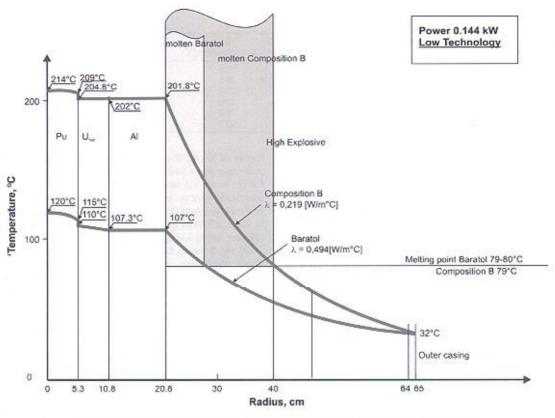


Fig. 3. Temperature profiles in a low technology HNED with an alpha-particle heat power of 0.144 kW.

Fig. 4 is based on an alpha-particle heat power of 0.240 kW, corresponding to reactor-grade plutonium from reprocessed UOX LWR spent fuel with a burnup around 60 GWd/t (Kessler, 2005). Temperatures in this case would be so high that medium-technology

high explosives (see Table 4) would melt or even reach the temperature limit above which a chemical explosion would be initiated. Reactor-grade plutonium from reprocessed UOX or MOX spent LWR fuel with a burnup above 55 GWd/t (roughly 3.2% Pu-238 in the

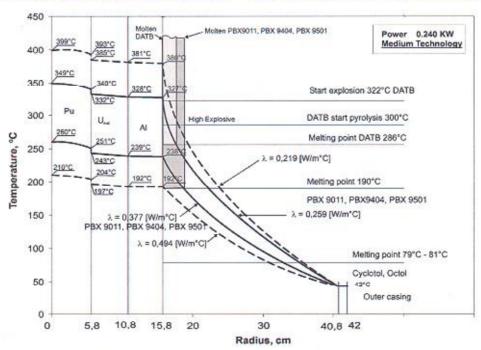


Fig. 4. Temperature profiles in a medium-technology HNED with an alpha-particle heat power of 0.240 kW.

Table 4
Characteristic materials data for medium technology high explosives

High explosive	Density (g/cm³)	Thermal conductivity (W/m°C)	Detonation velocity (km/s)	Melting point (°C)	Onset of pyrolysis (°C)	Temperature for onset of chemical explosion T _m (°C)	Thermal decomposition energy (kJ/kg)
DATB	1.83	0.259	7.6	286	300	322	1256
HMX	1.84	0.406	9.11	256-286	285	259	2094
PBX9011	1.77	0.381	8.5	190	260	?	2094
PBX 9404	1.84	0.385	8.8	190	290	236	2094
PBX 9407	1,65	0.335	8.4	204	240	?	2094
PBX 9501	1.86	0.452	8.7	190	275	235	2094

plutonium) would render a HNED based on medium technology technically unfeasible.

Fig. 4 also shows the temperature distributions for the Baratol and Composition B low-technology explosives of Section 3.2 as dotted lines. Note that the melting point for both explosives is 79–80 °C as shown in Table 3. Some 60–70% of the volume of the Baratol and Composition B high explosives would melt even if the explosive lenses were only 25-cm thick.

The results in Fig. 4 can confirm the statement in Fig. 1 only in part (for plutonium with less than 3.2% Pu-238, see above). However, it is not "technology comparable to the earliest plutonium weapons" but medium technology which was used in our calculations.

3.4. Additional remarks on the low and medium-technology cases

When assuming low technology qualifications no HNED will work if the available reactor-grade plutonium generates more heat than 0.120 kW. In case of an availability of medium technology qualifications the HNED will not function as long as the reactor-grade plutonium heat production stays above 0.240 kW. This is true even when neutronic analyses show nuclear explosion yields of about 0.12 kt TNT or 0.35 kt TNT, respectively (see Section 3.2).

Moreover, our thermal analysis is based on these conservative assumptions:

- Black-body radiation on the outer surface of the HNED casing (Kessler, 2007).
- If the radiation emissivities measured, e.g., for steel or aluminum at the outer casing were taken into account, the difference between the temperature of the outer casing and that of the ambient air (30°C) would increase by ε^{-1/4} (ε being the emissivity of the outer casing). For steel as the outer casing, ε = 0.22 (Paloposki et al., 2005; Kuechling, 1982). This leads to a temperature difference higher by a factor of 1.46. For aluminum, ε = 0.07 is reported, and a temperature difference higher by a factor of 1.94 is obtained.

In the opinion of the authors, the low-technology and mediumtechnology cases represent the highest levels of technology to be achieved in a NNWS. Subnational groups would not even be able to manage the low-technology case.

Yet, the high-technology case will be covered in the next section. This high-technology case can only be handled by some present NWSs after many years of research, experiments, and development.

3.5. High technology

This section contains a thermal analysis of a high-technology case (Fig. 5). Fig. 5 was already presented in (Kessler, 2007). The same HNED example was used by a US-Russian group for radiation analysis intended to detect hidden nuclear weapons (Fetter et al., 1990). In the authors' opinion, Fig. 5 is the most appropriate case for a high-technology HNED. This HNED has an outer diameter of 22 cm and a thickness of the spherical high explosive

lenses of 10 cm. The reactor-grade plutonium (density $15.8 \, \mathrm{g/cm^3}$) is arranged as a hollow spherical shell of 7.2 cm outer diameter and 5.6 cm inner diameter. Its mass is around 13 kg for $k_{\rm eff}$ = 0.98. Hollow fissile spheres were developed relatively early in the history of nuclear weapons (Cochran et al., 1987; Podwig, 2004). This design is more efficient and needs less volume for the implosion lenses (Kidder, 1974; Ashby, 1978; Seifritz, 1984).

As Fig. 5 shows, 0.858 kW of heat in the spherical shell of reactorgrade plutonium would exceed both the melting point and the temperature limit for chemical self-explosion of the high explosives. The TATB or PBX 9502 high explosives (see Table 5 and Fig. 5) have the highest thermal conductivity, the highest melting point, and the highest temperature for initiating chemical self-explosion to be found in the open scientific literature (Dobratz, 1972; Gibbs et al., 1981; Mader et al., 1982).

Even in such a high-technology HNED, with a heat power as low as 0.6 kW, the melting temperature would be exceeded by almost 20°C and the temperature for ignition of the TATB or PBX 9502 (see Table 5) high explosives would be exceeded by some 120°C. The temperatures at which chemical explosion starts are below the melting point for these two high explosives (Table 5). Again it must be emphasized that these calculated temperatures are conservative as the one-dimensional conservative approach and black-body radiation were assumed.

As 0.6 kW would correspond to a Pu-238 content of about 9% in the reactor-grade plutonium (Section 3.7), which exceeds Pu-238 contents in present UOX LWR or MOX LWR spent fuels of any burnup, it can be stated, as in Fig. 1 by the National Academy of Sciences (1995), that...plutonium of any quality produced in current nuclear power reactors...can be used to make nuclear explosives...

However, the authors' findings show this to be possible only with high technology. not as stated in Fig. 1... using technology comparable to the earliest plutonium weapons...

Only some NWSs (Biello, 2007) would be capable of mastering this high technology (geometric dimensions and type of chemical high explosives). Garwin (1998) points out that no NWS has ever used reactor-grade plutonium for nuclear weapons. This makes calculations and considerations of this kind absolutely hypothetical.

3.6. Coolability of HNEDs—how to handle the temperature limit problem

Statements in the literature asserted that the problem of temperature limits, e.g., melting temperatures or temperatures initiating explosion of high explosives, could be overcome by means of cooling (strips of conducting materials or external cooling) (Mark, 1993; Garwin, 1998).

3.6.1. Metal strips of high thermal conductivity

Garwin (1998) refers to the use of so called "in-flight insertion devices". In Nikitin (2007), preliminary analysis showed that a relatively large number of metal strips arranged symmetrically would have to be used for cooling. In addition, an insulating layer would

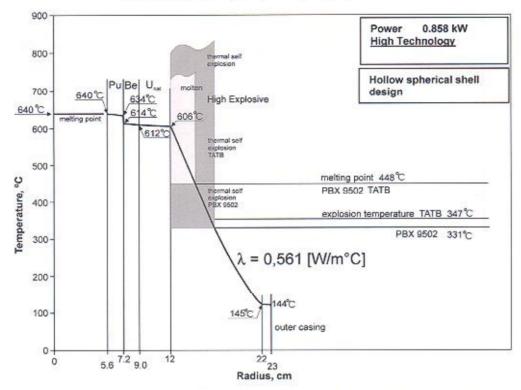


Fig. 5. Temperature profile in a high-technology HNED with a hollow spherical shell of reactor-grade plutonium in a design described by (Fetter et al., 1990). The adapted HNED with reactor-grade plutonium would produce 0.858 kW of heat.

have to separate the metal strips adjacent to the high explosives. Three-dimensional analysis would have to show the absence of azimuthal temperature increases between strips. Finally, the metal strips would have to be replaced by strips of high explosives assuring precise internal structures of the high explosive lenses before a HNED could be fired (otherwise detonation physics would not work). All of this would have to be done in a short span of time because interruption of cooling would raise temperatures in the HNED (Chen and Kessler, submitted).

3.6.2. Coolability of high technology HNEDs

Another idea for improving coolability was presented by (Shmelev et al., 2007). A high-technology HNED (Fig. 5) could be surrounded by a hollow spherical shell of aluminum 43-cm thick. The good thermal conductivity of aluminum and the larger outer surface would lower the outer temperature from 144°C (see Fig. 5) to something like 45°C. This would also lower the whole temperature profile of Fig. 5 by 99°C. Thus decreasing the lower temperature at the high explosive lenses from 606 to 507°C. Subsequent cooling by liquid nitrogen to -200°C would lower the whole temperature profile by another 245°C, resulting in 262°C for the inner temperature of the high explosive lenses. This is below the temperature at which self-explosion (331 or 347°C) of the TATB or PBX-9502 high explosives sets in (Table 5). Such HNED could still function. However, it would have to be assembled remotely under liquid nitrogen.

A hollow sphere with an outer radius of 7.2 cm at $k_{\rm eff}$ = 0.98 would have an outside temperature around 400 °C (at 30 °C outside air temperature). This gives an idea of some of the technical difficulties to be overcome.

There is no doubt that some NWSs (Biello, 2007) could overcome these technical difficulties. However, discussions of such hypothetical examples should stick to reality. HNEDs as shown in Fig. 5 are high-technology designs which could be built only by NWSs (geometric dimensions of high explosive lenses 10-cm thick and special high explosives developed in most advanced NWSs).

3.6.3. Effects of cooling low-technology HNEDs

Cooling a low-technology HNED shows a completely different result. Fig. 6 is a low-technology design with reactor-grade plutonium generating 0.24 kW of heat in its fissile part. The inner temperature of the high explosive lenses is 316 °C for the high explosive of lower thermal conductivity (Composition B), and 159 °C for the high explosive of higher thermal conductivity (Baratol) (Table 3).

If this low-technology HNED were cooled to $-200\,^{\circ}\text{C}$ by liquid nitrogen the entire temperature profile would be lowered by $234\,^{\circ}\text{C}$, the outer casing temperature in Fig. 6 being $34\,^{\circ}\text{C}$. The inner temperatures then would drop to $82\,^{\circ}\text{C}$ in the high explosive of lower thermal conductivity. Accordingly, the high explosive would melt on its inner surface (melting temperature $79\,^{\circ}\text{C}$) and disintegrate even if the low-technology HNED would be cooled to $-200\,^{\circ}\text{C}$.

Table 5
Characteristic materials data for high technology high explosives

High explosive	Density (g/cm ³)	Thermal conductivity (W/m°C)	Detonation velocity (km/s)	Melting point (°C)	Onset of pyrolysis (°C)	Temperature of onset of chemical explosion T _m (°C)	Thermal decomposition energy (kJ/kg)
PBX 9502	1.89	0.561	7.6	448	395	331	2512
TATB	1.89	0.544	7.6	448	395	347	2512

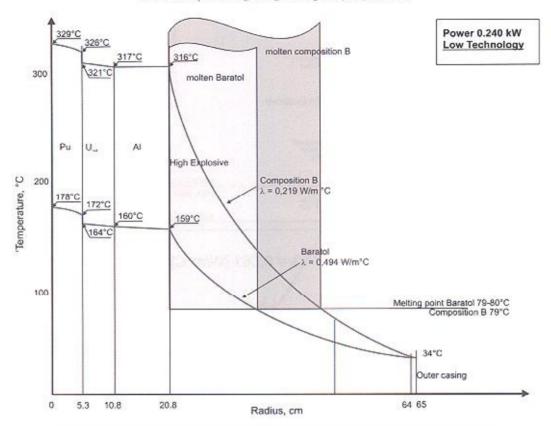


Fig. 6. Temperature profile in a low-technology HNED with reactor-grade plutonium of 0.240 kW heat output.

Submerging the HNED in liquid nitrogen has the consequence that the lower temperatures of all HNED materials give rise to lower thermal heat conductivities raising the above temperatures at the inner surface of the high explosives. This is shown by Chen and Kessler (submitted). Moreover, raising the heat power, e.g., to approximately 0.35 kW would cause a considerable volume of high explosives to melt and disintegrate.

3.6.4. Effects of cooling medium technology HNEDs

Fig. 7 shows a medium technology design with reactor-grade plutonium generating an assumed 0.614 kW in its fissile part. The inner temperature of 788 °C is indicated for medium technology high explosives of lower thermal conductivity, and 563 °C for medium-technology high explosives of the highest thermal conductivity.

If this medium-technology HNED were cooled to $-200\,^{\circ}\text{C}$ by liquid nitrogen, the temperature profile would be lowered by $262\,^{\circ}\text{C}$ the outer casing temperature being $62\,^{\circ}\text{C}$ in Fig. 7. The inner temperatures would then drop to $300\,^{\circ}\text{C}$ for the high explosive of higher thermal conductivity, and to $526\,^{\circ}\text{C}$ for the high explosive of lower thermal conductivity.

In a medium-technology HNED with approximately 0.42 kW heat power, the high explosives would still be molten or self-explode even if cooled to -200 °C by liquid nitrogen.

Again, as described in the previous section, temperatures of the high explosives will be higher due to the lower thermal conductivity at low temperatures (Chen and Kessler, submitted).

3.7. Conclusions drawn from neutronic and thermal analyses

It has become evident from previous sections that the results of neutronic analysis cannot be considered alone. For reactorgrade HNEDs, neutronic and thermal analyses must be seen together.

Thermal analysis leads to these results:

- · above 0.12 kW heat output of low-technology HNED designs;
- above 0.24kW heat output of medium-technology HNED designs:
- · above 0.60 kW heat output of high-technology HNED designs.

either the high-explosive lenses would partly melt and disintegrate or the temperature of the onset of chemical explosion would be exceeded. Such HNEDs would be technically unfeasible although neutronic analysis predicts that a minimum nuclear explosion fizzle yield could be attained as reported in Section 2. This is explained in Fig. 8.

If the HNEDs were cooled at the outside to -200°C by liquid nitrogen, these limits would rise to approximately 0.35 kW for low technology (Section 3.6.3), and to approximately 0.42 kW or slightly higher for medium technology (Section 3.6.4).

The Pu-238 content of the plutonium associated with this heat output is roughly

NNWS		
Low technology	0.12 kW	1.6% Pu-238
Medium technology	0.24 kW	3.2% Pu-238
NWS		
High technology	0.60 kW	9% Pu-238 (see Section 4 below)

In the light of the scientific and technical results outlined above, a lower limit of the heat output or Pu-238 content for purposes of proliferation resistance of reactor-grade plutonium can only be determined by IAEA by consent with the countries involved.

Fig. 7. Temperature profile in a medium-technology HNED with reactor-grade plutonium, heat power, 0.614 kW.

Radius, cm

4. Producing proliferation-resistant denatured plutonium

Reactor-grade plutonium with higher Pu-238 content is referred to as denatured and proliferation-resistant. Ways of producing this

material in various LWR fuel cycles are discussed below on the basis of data of Broeders and Kessler (2007). The Pu-238 content of low-enriched uranium (LEU) UOX fuel in current LWRs changes as a function of burnup, reaching approximately 3.7% Pu-

PBX9407, PBX9501

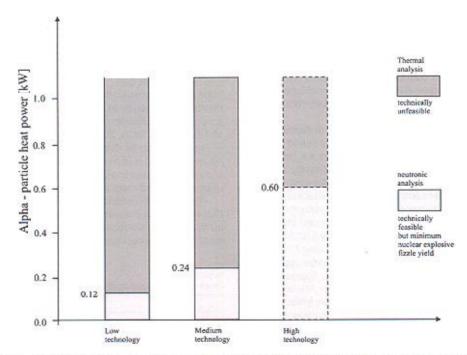


Fig. 8. Results of neutronic and thermal analyses, and limits above which HNEDs with reactor-grade plutonium become technically unfeasible for different levels of technology applied.

Table 6
Fuel compositions for producing denatured proliferation-resistant plutonium (Broeders and Kessler, 2007)

Fuel type	Pitch/Diameter P/D	Fuel composition	Th (wt%)	U (wt%)		Plutoniu	ım (wt%)	MA (wt%)
				Total	Fissile fraction	Total	Fissile fraction	
A	1.44	Re-enriched recycled U (RRU)	0	100	5.52	0	0	0
R	1.34	RRU U + Pu	0	93.9	5.00	6.1	64.5	0
D	1.41	RRU U + Pu + MA	0	92.5	5.00	6.5	64.5	1.0
E	1.59	Enriched U+Th+Pu+MA	52.6	35.1	6.00	10.7	64.5	1.6

Table 7

Composition (%) of proliferation-resistant plutonium from different types of fuel as a function of burnup in (Broeders and Kessler, 2007); subcritical masses and heat production of HNEDs based on such proliferation resistant plutonium

Fuel type Bu	Burnup (GWd/t)	Plutonium	Plutonium composition (%)					Subcritical mass	Total alpha-particle
		Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	heat (kW/kg)	$(kg)(k_{eff} = 0.98)$	heat power (kW)
Λ	50	9.5	50.5	21.7	13.2	5.1	0.0566	9.48	0.536
	58	11.4	46.1	22.5	13.4	6.6	0.0674	9.78	0.659
В	58	5.8	41	26.3	17.8	9.1	0.0358	10.61	0.380
	66	6.8	39.8	25.8	18	9.6	0.04148	10.64	0.441
D	49	9.6	39.3	27.2	15.7	8.2	0.0578	10.58	0.611
	58	10.7	37.4	26.8	16.1	9.0	0.0641	10.67	0.684
Е	49	8.9	33.7	32.9	15.0	9.5	0.0541	10.49	0.567
	58	10	30.6	33.3	15.8	10.3	0.0604	10.59	0.640

238 for burnups increased to 60 GWd/t. Higher Pu-238 contents can be achieved by using instead of LEU fuel, so-called enriched reprocessed uranium (ERU) or re-enriched reprocessed uranium (RRU) left over from reprocessing and then re-enriched. LWRs currently in operation have been fueled with this RRU (Gocke et al., 2006). Reprocessed uranium contains roughly 0.6–0.7% U-236 and 0.8–0.9% U-235. After re-enrichment, e.g., by centrifuge technology, this RRU can contain 3% U-236, 5.5% U-235, and 91.5% U-238. This RRU fuel is referred to as type A (Table 6) in this paper as in Broeders and Kessler (2007).

RRU can also be mixed with the reactor-grade plutonium from reprocessing spent LWR fuel with a burnup, e.g., of 50 GWd/t. This option is referred to as type B fuel (Table 6).

Reactor-grade plutonium can also be mixed with RRU and, in addition, with 1% minor actinides (MAs in the proportion of 49.2% neptunium, 46.7% americium, and 4.1% curium), generated in LWR fuel after a burnup of 50 GWd/t). This is referred to as type-D fuel (Table 6). Part of the U-238 contained in this type-D fuel can be replaced by Th. 232. This is type-E fuel in Table 6. In addition, type-E fuel contains 1.6% minor actinides of the isotopic composition described above.

These types of fuel were analyzed thoroughly in Broeders and Kessler (2007). For instance, the P/D ratio was determined in order to ascertain that all safety parameters of LWR cores, e.g., regarding coolant density reactivity effects, would fulfill licensing requirements. In addition, burnup calculations were performed to show the development of plutonium composition as a function of burnup. Table 7 shows the plutonium compositions after burnups of 49, 50 or 58 GWd/t. A Pu-238 content of 9.5% can be attained for type-A fuel already at 50 GWd/t. The Pu-238 content is only 5.8% for type-B fuel at a burnup of 58 GWd/t. For type-D fuel containing 1% MAs the Pu-238 composition would be 9.6% at 49 GWd/t, and for Type-E fuel it would be 8.9% Pu-238 at a burnup of 49 GWd/t. Approximately 2.5% or 3% U-233 would be produced in addition. These Pu-238 compositions, of course, can be modified to achieve lower Pu-238 percentages by choosing different initial fuel compositions.

Table 7 also shows the heat outputs of various HNEDs as a function of the plutonium compositions discussed above. The plutonium isotopes generating heat are shown in Table 8 corrected according to Chebeskov (2007) in comparison to values used for Pu-

242 in Kessler (2007). The plutonium compositions of type-A, D and E fuels would satisfy even the requirements for high-technology HNEDs with 0.6-kW heat output. Type-B fuel would only meet requirements for the medium and high-technology options with heat outputs of 0.38 kW and approximately 0.44 kW.

5. Strategy for a proliferation-resistant plutonium fuel cycle

The next issue to be discussed is the transition from present spent fuel to a denatured-plutonium (proliferation-resistant) fuel cycle by applying future advanced fuel cycle technologies.

Type-A fuel with RRU fuel would directly lead to denatured plutonium (Table 7 and Fig. 9). According to Fukuda (2004), present commercial uses of nuclear energy in the world will have produced ~2500 t of reactor-grade plutonium by 2010, ~80% of which will be held in interim storage while ~20% will have been reprocessed and held in MOX fuel (mainly in Europe and Japan) (Gocke et al., 2006). This reactor-grade plutonium, together with the associated neptunium, americium, and curium, can be incinerated by multiple recycling (Kessler, 2002; Broeders, 1992; Finck, 2005; Wade and Hill, 1997; Saito, 2005) in reactors instead of direct disposal of the spent fuel in deep repositories. In the long run the latter approach would lead to accumulation of all plutonium, neptunium, and americium and to a considerable additional future requirement of waste disposal capacity (Wigeland et al., 2005).

The use of RRU would require no change in present fuel fabrication technology, Present experience in Europe and an assessment by the Argonne National Laboratory (Finck, 2005) show fuel cycle costs for plutonium recycling (present MOX fuel or metal fuel) and

Table 8
Characteristic heat outputs of various Pu isotopes

	α-heat output (W/kg)
Pu-238	570
Pu-239	1.9
Pu-240	6.8
Pu-241	3.3
Pu-242	0.15

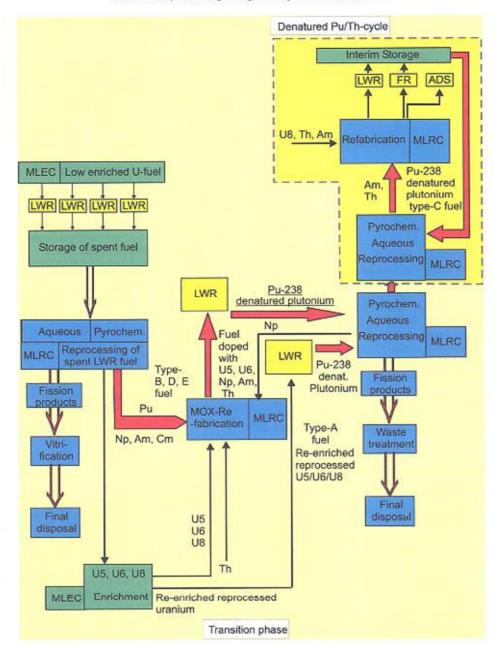


Fig. 9. Transition from present interim storage of spent UWR fuel to reprocessing and refabrication (in multilateral enrichment centers (MLEC) and multilateral reprocessing/refabrication centers (MLRC)) of dedicated fuel to a civil denatured-plutonium fuel cycle with incineration of plutonium as well as minor actinides.

waste management to be a few percent higher than for present LWR LEU fuel with burnups of 50–60 GWd/t and subsequent interim and deep geologic storage (direct fuel disposal). Extensive experience in refabrication and irradiation of present MOX fuel up to a burnup of 50–55 GWd/t is available mainly in Europe (Krellmann, 1993; Picard et al., 2000; Gocke et al., 2006).

As Fig. 9 shows, the spent fuel at present kept in interim storage would have to be reprocessed by future advanced aqueous or pyrochemical techniques (Herbig et al., 1993; Hill et al., 1995; Laidler et al., 1997; Finck, 2005; Conocar et al., 2006). At present, only aqueous reprocessing technology (PUREX) is available on a technical scale at La Hague, France, Sellafield, United Kingdom, and Tokai-mura, Japan. The reactor-grade plutonium and MAs (neptunium, americium, curium) separated together with uranium and thorium would

have to be fabricated into type-B, D and E fuels in a future refabrication plant for fueling LWR cores. If fuel doped with americium and curium became too difficult to refabricate, both americium and curium could be separated, only americium could be irradiated, and curium could be stored.

IWR core design would need some minor changes and adaptations to the control rod systems, e.g., more control rods, more liquid poison, and poison rods with highly-enriched boron (Barbrault, 1996; Aniel-Buchheit et al., 1999). Design possibilities for full MOX cores were suggested by (Barbrault, 1996). Higher P/D ratios or water rods would entail lower electric power outputs or larger diameter of the pressure vessel.

After a burnup of 50-60 GWd/t, plutonium will be converted into proliferation-resistant denatured reactor plutonium as shown

Table 9
Plutonium composition of denatured proliferation-resistant type-C plutonium fuel after one burnup cycle

Isotope	Initial loading (wt%)	Burnup (GW	/d/t)
		49	58
Pu-238	7.7	8.4	8.8
Pu-239	44.0	37.0	36.4
Pu-240	31.0	27.4	26.7
Pu-241	10.3	17.6	17.8
Pu-242	7.0	9.6	10.3

above for type-A, B, D and E fuels. Type-D and E fuels containing MAs need further development of fuel refabrication technologies.

The higher Pu-238 content of converted denatured proliferation-resistant reactor plutonium produces more heat and spontaneous neutron radiation and γ-radiation. Present aqueous reprocessing technology would have to be modified accordingly, and present MOX refabrication technology would no longer be feasible. Present glove-box-type MOX refabrication technology is limited to a Pu-238 content of ~4%, present aqueous reprocessing technology, to ~5%. Advanced aqueous and pyrochemical reprocessing and related refabrication technologies for metallic fuel and MOX fuel would have to be developed and remote handling would become mandatory. The advanced technologies currently under development in actinide transmutation research in the United States, Russia, Japan, and Europe would have to be employed (Herbig et al., 1993; Laidler et al., 1997; Conocar et al., 2006).

Penalties in fuel cycle costs, compared to present MOX fuel, certainly will have to be paid as a function of the expense required in advanced reprocessing and refabrication technologies for type-A and B as well as D and E fuels.

Long-term behavior of denatured fuel in light water reactors and fast reactors

Once denatured reactor plutonium would be introduced into the denatured-plutonium fuel cycle it could be incinerated by multiple recycling in LWRs or FRs, either in integrated fast reactors (IFRs) (Hill et al., 1995; Wade and Hill, 1997) or FRs of the Consommation Améliorée du Plutonium dans les Réacteurs Avancés (CAPRA) type (Tommasi et al., 1994; Languille et al., 1995; Messaoudi et al., 2002). Recycle time for incineration of denatured reactor plutonium should be less than a few decades, for Pu-238 has an α -decay halflife of 87.7 years. Following this multirecycling strategy gives rise to the question how the Pu-isotopic composition would change during irradiation to a certain burnup in these reactors with different neutron energy spectra (LWRs, FRs and ADSs).

6.1. Long-term behavior of denatured plutonium in LWRs

MOX fuel containing denatured reactor plutonium and RRU can be loaded into a PWR core with the pin-cell parameters shown in Table 6. This is referred to as type-C fuel (Table 9). The Pu-238 fraction of this type-C fuel (denatured reactor plutonium) increases slightly from 7.7 to 8.4% or 8.8% after burnups of 49 or 58 GWd/t, respectively, in a PWR core. In addition, some 250 kg of denatured plutonium is incinerated in a 1 GWe PWR over a burnup cycle of 60 GWd/t (Broeders and Kessler, 2007). As shown in Broeders (1996) and Kessler (2002), reactor-grade plutonium should not be recycled more than three times in LWRs. Multiple recycling of denatured plutonium in LWRs is not possible, as will be explained in Section 8 below.

6.2. Long-term irradiation behavior of denatured plutonium in fast reactors

MOX fuel with denatured reactor plutonium mixed with natural uranium loaded in a typical FR core decreases the Pu-238 isotope concentration as a function of burnup because of the higher fission/absorption ratio in the fast neutron spectrum (Wade and Hill, 1997). However, over a typical possible burnup of 150 GWd/t in a typical FR core, this is only a net decrease of approximately 2% of the Pu-238 content (Broeders and Kessler, 2007). A Pu-238 content raised, e.g., to some 11% at the beginning of the burnup cycle would decrease to approximately 9% by the end of the burnup cycle. The reactor plutonium thus would remain denatured throughout the burnup cycle and after unloading of the fuel. FRs, because of their more efficient destruction of Pu-238, would permanently need a certain supply of the Pu-238 isotope either from LWRs or by slightly modifying the FR fuel so that the Pu-238 content stays constant over the burnup cycle (see Section 8).

There is a problem, however, if neptunium and americium are used for the production of Pu-238 in denatured proliferation-resistant plutonium. The neptunium minor actinide is an isotope susceptible to proliferation like U-235 or U-233. This aspect will be discussed in the next chapter. Americium can be shown to be proliferation-resistant (Kessler, 2008), as will also be discussed below. Consequently, the neptunium minor actinide should be used only in the transition phase (see Fig. 9) to produce denatured proliferation-resistant plutonium. This could be achieved best in reprocessing plants of present NWSs, e.g., La Hague, France and Sellafield, UK during the transition phase, or in multilateral fuel cycle centers (IAEA, 2005). Later, when proliferation-resistant denatured reactor-grade plutonium will be used in the civil proliferation-resistant fuel cycle (Fig. 9) neptunium must be avoided.

7. Minor actinides

The neptunium, americium, and curium minor actinides are part of nuclear waste in LEU-UOX and MOX spent fuel elements. They are used in type-D and E fuel scenarios (Section 4) to produce denatured proliferation-resistant plutonium. They would be produced also in spent type-A, B and C fuel scenarios (Section 4).

These minor actinides also have low critical masses (Seifritz, 1984). They therefore must be discussed in the light of proliferation and IAEA safeguards. Solving the proliferation problem of reactorgrade plutonium by producing denatured proliferation-resistant plutonium (Sections 2–6) is not sufficient. The proliferation problem of minor actinides must be solved as well.

7.1. Neptunium

Neptunium is considered usable in nuclear explosive devices (Loaiza et al., 2004, 2006). It has a bare critical mass of $57 \pm 4 \,\mathrm{kg}$ (Sanchez et al., 2008). A reflector, e.g., beryllium can reduce this bare critical mass to approximately $45 \,\mathrm{kg}$. It produces no alpha heat and has a very low spontaneous fission neutron rate of $0.11 \,\mathrm{n/kg} \,\mathrm{s}$, which is as low as for U-235 (0.29 $\mathrm{n/kg} \,\mathrm{s}$) (Holden et al., 2000). The IAEA has begun to adopt measures to monitor neptunium (Albright et al., 1999; Ottmar et al., 2001; Morgenstern et al., 2002). The amount of neptunium available in civil nuclear energy programs is estimated by IAEA to be around $90 \,\mathrm{t}$ (Fukuda et al., 2006). Neptunium, therefore, should be incinerated already in the transition phase (Fig. 9) by the NWSs and should be avoided as far as possible in the following denatured-plutonium fuel cycle (Fig. 9). Suggestions for such a fuel cycle strategy avoiding neptunium will be discussed in Section 8 below.

7.2. Americium

Americium generated in nuclear reactors is a mixture of Am-241, Am-242m, and Am-243. Am-241 without admixture of other americium isotopes can also originate from the decay of Pu-241. The critical mass of Am-241 was calculated to be approximately 34-45 kg, that of Am-243, between 111 and 193 kg, in both cases with steel as reflectors (Diaz et al., 2003).

The critical mass of Am-242m is as low as 3.7-5.6 kg when reflected by steel (Diaz et al., 2003). However, Am-242m amounts to less than 1% in the spent LWR fuel, and approximately 4% in spent fuel of FRs or ADSs, as was shown in a detailed analysis in Kessler (2008). Spontaneous fission neutron emission of Am-241, Am-242m and Am-243 is relatively high. Using reactor-grade americium as a fissile material in gun-type HNEDs would lead to only negligible nuclear explosive fizzle yields (Kessler, 2008). Reactor-grade americium thus could only be used in implosiontype HNEDs, Nuclear explosion yield considerations result in only minimum nuclear explosion fizzle yields. These are similar to those for reactor-grade plutonium. However, Am-241 has a high alpha particle heat output (110 W/kg). The subcritical mass for k_{eff} = 0.98 of reactor-grade americium would be in the range of 48-58 kg. This leads to alpha particle heat outputs of more than 3 kW for HNEDs based on reactor americium. Thermal analyses show the melting temperatures and temperatures for the onset of chemical selfexplosion in the high explosive lenses to be exceeded. HNEDs based on reactor-grade americium as the fissile material would be technically unfeasible although non-negligible nuclear fizzle explosive yields can be calculated under the assumption that the implosion technique will be used (Kessler, 2008).

This was shown in an extensive analysis of Am-241 originating from the decay of Pu-241 as well as for various reactor-grade mixtures of Am-241, Am-242m, and Am-243 generated in various LWR, FR and ADS fuel cycles (Kessler, 2008).

7.3. Curium

Curium isotopes have a relatively low critical mass, but a high spontaneous fission neutron rate and a very high alpha heat production within a critical mass. They can, therefore, be excluded from proliferation discussions (Seifritz, 1984).

8. Neptunium-free nuclear fuel cycle

Sections 5 and 6 above showed that reactor-grade plutonium can be converted so as to become denatured and proliferationresistant. Kessler (2002) demonstrated that such reactor-grade plutonium can also be incinerated (even completely, if required). This also holds for the incineration potential of the neptunium and americium minor actinides. Minor actinide incineration is being discussed also with a view to minimizing the radioactive inventory of nuclear waste disposal sites (Kessler, 2002; Wigeland et al., 2005). Neptunium, however, poses a problem because of its usability in nuclear explosive devices. Section 6, therefore, proposed to use it only in the early transition phase (Fig. 9) for doping, e.g., types-D and E fuels, and to reprocess or refabricate such fuel only in fuel cycle centers of NWSs. Neptunium should be avoided in the following phases of a civil denatured fuel cycle, while denatured proliferation-resistant plutonium and reactorgrade americium should be incinerated.

8.1. Model of a neptunium-free nuclear fuel cycle

The neptunium actinide cannot be denatured with other neptunium isotopes. In addition, it is considered a fissile material usable

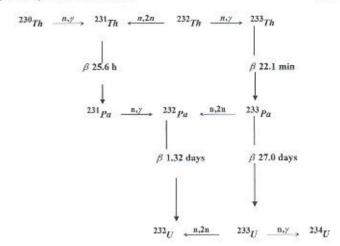


Fig. 10. Buildup of isotopes in the thorium fuel cycle.

in nuclear explosives (Section 7.1). Therefore, it must be avoided in a future denatured proliferation resistant civil nuclear fuel cycle.

Galperin et al. (2000) and Sagara (2006) offer some indications that this is possible by combining denatured proliferation-resistant plutonium (Section 4) with depleted U-238 and thorium. In this way, neptunium can be avoided as only protactinium isotopes and uranium isotopes up to U-234 (Fig. 10) or plutonium isotopes and the americium and curium minor (Fig. 11), but no significant amounts of U-236 or neptunium are produced. In addition, americium can be used which would produce some Pu-238. This occurs by buildup and alpha decay of Cm-242 (Fig. 11). Irradiation experiments in a fast reactor have shown that roughly 2% Pu-238 can be produced by converting 2–3% americium into Cm-242 (Walker et al., 1995). The use of thorium in addition allows new U-233 to be produced.

A reactor core would need fissile and fertile fuel rods. The fissile fuel rods would contain denatured proliferation-resistant plutonium (Sections 4 and 5) and depleted uranium (as little U-235 as possible) from enrichment plants. Denatured proliferation-resistant plutonium would be incinerated as shown in (Broeders and Kessler, 2007). U-238 would generate Pu-239, thus improving the isotopic composition of denatured proliferation-resistant plutonium. In addition, some americium (either homogeneously distributed in the fuel or in special fuel rods) would be added to generate Pu-238 in case the Pu-238 content decreased during burnup.

The fertile fuel rods would contain thorium, some U-238 in the appropriate proportions (roughly 90% thorium, 9.5% U-238) to generate denatured uranium with <12% U-233. As U-238 would generate some Pu-239, a small percentage, e.g., 0.5%, to several % of denatured proliferation-resistant plutonium could be added. In this way, the newly generated Pu-239 would be incorporated into the denatured proliferation-resistant plutonium.

In a future civil denatured-fuel cycle (Fig. 9), this would allow both the denatured proliferation-resistant plutonium and the americium to be incinerated. It also would generate some U-233 from thorium. It would work in LWRs, ADSs and FRs. All fuel would be proliferation-resistant, and only IAEA safeguards of the kind required for LEU-LIOX LWR fuel would have to be applied.

Multiple recycling of reactor-grade plutonium or denatured proliferation-resistant plutonium is not possible in LWRs because of the changes in plutonium composition. This causes the void coefficient, one of the key safety factors to turn positive after some three full recycling phases. Also, the amount of americium produced increases tremendously as a function of each recycling step

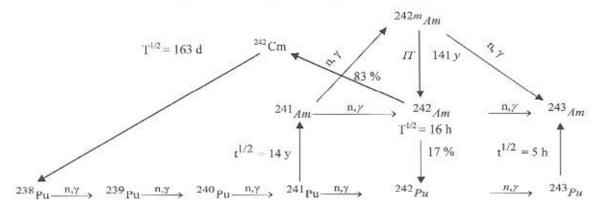


Fig. 11. Production routes of americium and curium isotopes, and α-decay of Cm-242 to Pu-238.

(Broeders, 1992, 1996). Three recycling phases with fresh plutonium from other LEU-UOX LWRs added in each cycle would extend over a period of 70 years. in which roughly one half of the plutonium produced would be incinerated. More than three recycling phases would produce too much Cf-252 (Salvatores, 2008), which would prohibit fuel refabrication if actinides are not separated.

After some two full recycling phases plutonium in LWR-MOX reactors would be loaded in FRs and, in special cases, fast-spectrum ADSs where multirecycling is possible because of the more favorable fission/absorption ratio in the fast neutron spectrum (Rineiski and Kessler, submitted for publication).

9. Conclusion

The authors' analysis and assessment of the proliferation risk of reactor-grade plutonium can be summarized as follows:

- · Neutronic analysis of HNEDs with reactor-grade plutonium as the fissile part of spherical implosion-type devices indicates early neutronic pre-ignition (about 1 µs after prompt criticality) and nuclear explosive yields of 0.12 kt TNT (equivalent) for the low technology and 0.35 kt TNT (equivalent) for the very high technology scenarios. Analysis of the probability density function for pre-ignition shows this to be very narrow. The nuclear energy yields releases thus become quasi-deterministic. These values are somewhat below the 0.54kt TNT (equivalent) reported by Mark (1993). The authors' results are smaller quantitatively but not different qualitatively. This nuclear explosive yield potential of reactor-grade plutonium, which is based on the assumption of perfect radial symmetry of the shock wave applied to the outer surface of the reflector, still exceeds the explosive yield of any known conventional chemical explosive. Besides the nuclear explosive yields, also the radiation and radiotoxicity effects of such reactor-grade plutonium nuclear explosions would have to be taken into account. These results of neutronic analysis however, are only part of the reality.
- Thermal analysis shows a different picture under assumptions about the geometric dimensions and the level of technology needed for the type of chemical explosives and the thickness of high explosive lenses.
- Low-technology HNEDs with reactor-grade plutonium and the type of high explosives or geometric dimensions used in the earliest nuclear weapons are not feasible technically. Their high explosive lenses would melt and disintegrate. This is valid for all reactor-grade plutonium from reprocessing LEU-UOX or MOX spent fuels with a burnup of more than 30 GWd/t. Alpha-particle

- heat above 0.12 kW would render such HNEDs unfeasible technically.
- Medium-technology HNEDs with high explosives classified as medium technology (an account of their thermal conductivity, melting point and temperature for the onset of chemical explosion) and chemical explosive lenses 25-cm thick would be unfeasible technically for reactor-grade plutonium from reprocessing LEU-UOX and MOX LWR spent fuel with burnups of more than 55 GWd/t. This corresponds to a heat output of reactor-grade plutonium in a HNED of more than 0.240 kW. For reactor-grade plutonium producing less than 0.24 kW, medium-technology HNEDs would be feasible technically.
- The authors conclude that NNWSs could only master low technology and, to some extent, perhaps medium technology.
 Subnational groups could not even handle low technology.
- Assuming high technology for the geometric dimensions (hollow reactor-grade plutonium sphere, high explosive lenses 10-cm thick) and for the chemical high explosives (highest thermal conductivity, highest melting point and highest temperature for the onset of chemical explosion), which is available in NWSs, this limit increases to approximately 0.6 kW, corresponding to some 9% Pu-238 in reactor-grade plutonium. High-technology HNEDs of NWSs would be unfeasible technically with reactor-grade plutonium containing more than approximately 9% Pu-238 or generating more than 0.6 kW. For reactor-grade plutonium containing less than 9% Pu-238 high-technology HNEDs would be feasible technically.
- The statement by the US National Academy of Sciences (1995)
 (Fig. 1) can be confirmed for plutonium from nuclear reactors
 with up to 9% Pu-238 if high technology is used, which is only
 available in NWSs. The nuclear explosive yield attainable would
 still be in the range shown by the neutronic analyses described
 in this paper.
- Remote assembly of the HNED in a pool of liquid nitrogen for cooling is discussed in Section 3.6. If these extremely difficult technical operations could be mastered, the lower limits indicated above, for which reactor-grade plutonium based HNEDs would not be technically feasible would change:
 - for a low-technology HNED, from 0.12 kW to approximately 0.35 kW.
 - for a medium-technology HNED, from 0.24 kW to slightly more than 0.42 kW.
- However, technical facilities only available to NWSs should not be mixed up with proliferation by NNWSs. The authors find the appropriate limit for proliferation resistance of reactor-grade plutonium eventually can only be determined by IAEA in consent with the countries involved.

Section 4 presents four fuel cycle options (see Table 6) leading to sufficiently high Pu-238 percentages in spent fuel. Type-A fuel contains RRU in the fresh fuel elements and would lead to denatured proliferation-resistant plutonium with more than 9% Pu-238 after a burnup around 50 GWd/t. Type-B fuel contains plutonium separated from present spent LEU-UOX LWR fuel with a burnup of 50 GWd/t and additional RRU. It would lead to some 6% Pu-238 in denatured proliferation-resistant plutonium after a burnup of 60 GWd/t. Type-D fuel contains plutonium and RRU as type-B fuel plus some 1% minor actinides. It would produce denatured proliferation-resistant plutonium with approximately 11% Pu-238 after a burnup of 60 GWd/t. In type-E fuel, some of the uranium in type-D fuel would be replaced by thorium. This again would lead to denatured proliferation resistant-plutonium with 11% Pu-238 and approximately 3% U-233 after a burnup of 60 GWd/t. All these Pu-238 fractions would give rise to a heat output of 0.38 to more than 0.68 kW in the HNEDs. All these high Pu-238 shares could be adjusted to lower percentages by adapting the initial fuel compositions accordingly.

Section 5 discusses a transition phase in which all plutonium now in existence could be converted into denatured proliferation-resistant plutonium. Also type-A fuel (RRU) would directly produce denatured proliferation-resistant plutonium but also neptunium. RRU can be enriched in existing enrichment centers. Existing reprocessing centers could be expanded by reprocessing centers, preferably in NWSs, in the near future. Conversion of present reactor-grade plutonium is only a matter of reprocessing and refabrication capacity, since refabrication of the fuel and burnup of the type-A, B, D and E fuels and reprocessing of the spent fuel takes only about 12 years.

Denatured proliferation-resistant plutonium would only be subject to IAEA safeguards of the type now required for LEU-UOX LWR fuel or <12% U-233 denatured uranium. This denatured reactorgrade plutonium could then be incinerated in a civil denatured proliferation-resistant fuel cycle.

Present IAEA safeguards of plutonium must remain in place for reactor-grade plutonium producing heat in HNEDs below the limits discussed in the low and medium-technology scenarios.

An additional proliferation problem arises for the neptunium and americium minor actinides. Reactor americium can be considered proliferation-resistant as it appears in all fuel cycle strategies as a mixture of Am-241, Am-242m, and Am-243. Also Am-241, which originates from the decay of Pu-241, can be considered proliferation-resistant. This was shown by (Kessler, 2008). The neptunium minor actinide can be used in nuclear weapons and cannot be denatured. Proposals to keep minor actinides together with plutonium in fuels such as type-D or E are no solution yet of the proliferation problem. Neptunium can be separated chemically like reactor-grade plutonium. This should be done only in the transition phase, preferably in reprocessing centers of NWSs or MLRCs (see Fig. 9). In the subsequent phase of a civil denatured proliferation-resistant fuel cycle, neptunium should be avoided. A fuel strategy avoiding neptunium and allowing denatured proliferation-resistant plutonium and americium to be incinerated is proposed in Section 8. Reprocessing centers in the civil denatured proliferation-resistant fuel cycle should be multilateral reprocessing centers as recommended by IAEA.

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