PROLIFERATION RISK RELATED TO THE DIVERSION OF REACTOR-GRADE PLUTONIUM

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ABSTRACT – *The reactor-grade plutonium produced in the fuel of a typical LWR, at the time of discharge, is generally considered no-attractive for the realization of nuclear weapons, due to the presence of ^{240}Pu in high concentration (a strong neutron emitter due to its spontaneous fission) and ^{238}Pu (a strong α-emitter). However, in the last decades, doubts on the possible use of reactor-grade plutonium for nuclear weapons have arisen. It is, therefore, important to understand how reactor-grade plutonium, from nuclear fuel cycle, can be potentially used to produce nuclear explosives. Aim of this work is to evaluate, making use of available literature data and physical models, what is the potential nuclear explosive yield of an Hypothetical Nuclear Explosive Device (HNED) of the implosion type, based on the reactor-grade plutonium and low technology, i.e. a technology comparable to that of the first plutonium weapons. The results of the numerical analysis shows that the high inherent spontaneous fission neutron source of ^{240}Pu, strongly affects the explosion yield, so limiting the energy released to the range of fraction of kilotons (TNT equivalent).*

Keywords: Non-proliferation, plutonium, nuclear fuels, safety

1. Introduction

It is well known that the fissile plutonium ^{239}Pu, as well as the higher order isotopes ^{240}Pu, ^{241}Pu and ^{242}Pu, are produced in the reactors by successive neutron capture of ^{238}U isotope. In addition, another isotope of plutonium, ^{238}Pu, is accumulated in the fuel as consequence of neutron capture of ^{237}Np. The concentration of these plutonium isotopes, in the fuel, is nearly related to the time of irradiation in the reactor: the longer is the irradiation time, the higher is the concentration of ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu. On the other side, plutonium can be used in Light Water Reactors (LWRs) by reprocessing of spent fuel, in the form uranium-plutonium mixed oxide (MOX). It is clear that, even in different way, the concentration of plutonium in the irradiated fuel, as well as its re-use as MOX, poses concerns for its potential diversion and use for non-peaceful applications. The reactor-grade plutonium generated in LWRs, after a typical burn-up of 60 GWd/t, is generally considered no-attractive for the realization of nuclear weapons due to the presence, in high concentration, of ^{240}Pu (a strong neutron emitter for spontaneous fission), ^{238}Pu (a strong α-emitter), as well as of the high radiation level from ^{241}Am. For all these reasons, the production of plutonium usable for weapon application (weapon-grade or super-grade plutonium), requires that the fuel is irradiated in ad-hoc reactors and facilities for a very short time (few GWd/t), to minimize the concentration of these isotopes. Nevertheless, in the last decades, several qualified experts highlighted the potential proliferation concerns related to the use of reactor-grade plutonium. These facts were also recognized by the International Atomic Energy Agency (IAEA), from its beginning, where all plutonium, except

plutonium with an isotopic concentration of ^{238}Pu greater than 80%, is regarded as equally hazardous from the point of view of diversion to nuclear weapon (IAEA, 2002). It is, therefore, important to understand how reactor-grade plutonium, from nuclear fuel cycle, typically containing 50-60% of fissile (compared with 93% of fissile for weapon-grade material), can be potentially used to develop a nuclear weapon, even with low yield but having devastating effects. Aim of this work is to evaluate, making use of available literature data and physical models, what is the potential nuclear explosive yield of an Hypothetical Nuclear Explosive Device (HNED)[a] of the implosion type, based on the reactor-grade plutonium and low technology, i.e. a technology comparable to that of the first plutonium weapons. To this end, an established methodology was used while employing new codes and data libraries.

2. Plutonium mixtures and material security

2.1 Classification of plutonium grades

Depending on its isotopic composition, plutonium can be classified as super-grade (SG), weapon-grade (WG), fuel-grade (FG) and reactor-grade (RG), the last one being produced in the commercial nuclear power reactors. Typical values of the isotopic concentrations (in weight fraction) for various plutonium grades, are reported in the Table 1. The weapon-grade is the standard material for nuclear weapon; it is easy to use, with high yield, low radiation level and low heat generation. The super-grade is even better. Plutonium with an isotopic content of ^{240}Pu from 7% to 18% is referred as fuel-grade, and up to seventies, nobody saw any interest in this fuel for serious weapon use, due to the higher radiation and heat levels. However, in 1977, the Department of Energy (DOE) announced that in 1962, USA exploded a device using fuel-grade plutonium supplied by United Kingdom. That successful test, with a yield less than 20 kiloton[b] (TNT equivalent), confirmed that plutonium, with grade different from that traditionally considered in the weapon manufacturing, in combination with an adequate technology, could be used to make a nuclear explosive (Pellaud, 2002).

Table 1 - Typical isotopic concentrations for various plutonium grades (Pellaud, 2002), (Mark,1993), (Kessler, 2011).

Grade	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
SG	-	0.98	0.2	-	-
WG	1.2×10^{-4}	0.938	0.058	3.5×10^{-3}	2.2×10^{-4}
FG	$\approx$ 7% – 18% ^{240}Pu				
RG(I)	0.038	0.518	0.231	0.142	0.071
RG(II)	0.055	0.341	0.311	0.106	0.187

As far as reactor-grade plutonium, two different compositions are considered in Table 1. The first one, named RG(I), is a typical plutonium isotopic composition of LWR spent fuel, after 60 GWd/t exposure; the second one, labelled as RG(II), also referred as MOX-grade, represents an isotopic plutonium composition after three times recycling in MOX fuelled LWRs, and its content of ^{240}Pu is so high that it is considered proliferation-proof (Kessler, 2011). However, reactor-grade plutonium is theoretically usable to make a crude explosive device (Pellaud, 2002).

2.2 Property of plutonium isotopes

It is known that all plutonium isotopes are fissile for fast neutrons; this means that a bare critical assembly, i.e. a critical assembly obtained without the use of any neutron

[a] The term HNED is referred to a device which is not based on any real design.

[b] 1 kT TNT equivalent = 4.187×10^{12} J

reflector, could be made with plutonium metal, no matter what is its isotopic composition. The values of critical mass M_c (bare-sphere), spontaneous (fission) neutron emission rate S, and power density P_α due to α-decay, for the isotopes ^{238}Pu, ^{239}Pu, ^{240}Pu , ^{241}Pu and ^{242}Pu, in δ-phase[c], are given in the Table 2 (Kessler, 2011),(Kimura, 2012). It can be demonstrated that, at all burn-up levels and at any time following the fuel discharge, the critical mass of reactor-grade plutonium is intermediate between that of ^{239}Pu and ^{240}Pu (Mark, 1993).

Table 2 - Properties of the plutonium isotopes.

Parameter	**^{238}Pu**	**^{239}Pu**	**^{240}Pu**	**^{241}Pu**	**^{242}Pu**
M_c (kg)	13.1	14.8	44.8	17.6	87.8
S (n/kg/s)	2.6×10^6	20	9.1×10^5	50	1.7×10^6
P_α (W/kg)	570	1.9	6.8	3.3	0.15

The heat produced by α-decay of ^{238}Pu can pose serious technical obstacles to the realization of an HNED of the implosion type, due the impact of the decay heat on the stability of explosive lenses, i.e. the chemical explosives used to generate the converging implosion shock waves, that make the plutonium core to implode. Several authors (Kessler, 2001),(Kimura, 2011), have shown that the use of plutonium containing more than 15% of ^{238}Pu, could make technically unfeasible the development of such HNED using an early technology, i.e. a technology comparable with that available at time of realization of the first nuclear weapons. Another important nuclear property affecting the manufacture of an HNED is the spontaneous fission neutron emission, due to some plutonium isotopes. In fact, strong inherent neutron emission may cause premature initiation of the chain reaction, before the plutonium is fully compressed, and maximum reactivity is inserted in the fissile material. This is known as "pre-detonation", and it reduces the nuclear explosive yield. As reported in Table 2, neutrons from spontaneous fission are produced by plutonium isotopes with even mass number as ^{238}Pu, ^{240}Pu and ^{242}Pu. From the beginning of the nuclear age, proliferation issue was mainly focused on plutonium denaturing by increasing the isotope fraction of ^{240}Pu, being the plutonium with high content of ^{240}Pu already produced in typical LWR fuels. However, aside from ^{240}Pu, the other plutonium isotopes with even mass number, ^{238}Pu and ^{242}Pu, can also contribute to enhance the proliferation resistance of plutonium, thanks to their higher spontaneous fission neutron emission rate in comparison with ^{240}Pu.

3. Implosion process in an HNED

In an HNED of the implosion type, the typical core is a solid spherical mass of fissile material (uranium or plutonium highly enriched in the isotopes ^{235}U or ^{239}Pu), and the burst is activated by means of a strong positive reactivity insertion in the fissile material, following the compression. Chemical explosive in the form of lenses, surrounding the plutonium or uranium sphere, drives the implosion process. The physics describing the implosion process is very complex. In fact, during the compression phase, an inward shock wave propagates in the core, while the neutron density rises extremely fast after the transition to the prompt critical regime. The accumulating internal energy causes the melting of the fuel, its vaporization, then the vapour pressure build-up, and finally ionizes (plasma). The inward shock wave will progress toward the center of the core until the rising pressures will counteract it, and stop it. From that point on, the expansion phase will dominate the physics, with a quick disassembling of the system. A rigorous description of the physics would require the use of equations for the conservation of momentum,

[c] Plutonium metal has six allotropic forms, corresponding to six different crystalline configurations. The two forms most mentioned, with respect to weapons, are the α-phase (density = 19700 kg/m^3 at 340K), and δ-phase (density = 15800 kg/m^3 at 600K) (Kessler, 2011).

mass and energy, coupled with the equations describing the neutronics of the system. However, simple analytical models able to describe the physics of the implosion process and to simulate the nuclear explosion, providing a description of the timing of events involved in the excursion, have been developed (Dolan, 1982) (Seifritz 2009); these models will be herein used for the evaluation of the potential nuclear explosive yield of an HNED based on plutonium from fuel cycle .

3.1 Velocity of the imploding core

For the plutonium sphere undergoing the implosion process, it is possible to derive an analytical expression for the velocity of the particle u_p at the surface, as function of the applied external pressure p. The procedure here used is similar to that proposed by Dolan (Dolan, 1982), in the inertial confinement fusion, for the calculation of the radial velocity of a pellets that implodes under the effect of an ablation pressure. The starting point is the Newton's Second Law applied to the spherical core of radius R and mass m (assumed constant), where an external pressure p is applied on the surface:

$$\frac{d}{dt}\left(m\frac{dR}{dt}\right) = -4\pi R^2 p$$

Multiplying both sides by (2/m)(dR/dt), and assuming m = constant, the previous equation becomes:

$$2\frac{dR}{dt}\left(\frac{d^2R}{dt^2}\right) = -\frac{8\pi p R^2}{m}\frac{dR}{dt}$$

or

$$\frac{d}{dt}\left(\frac{dR}{dt}\right)^2 = -\frac{8\pi p R^2}{m}\frac{dR}{dt}$$

with the initial conditions: R = R_o and dR/dt = 0 for t = 0. In the hypothesis that the pressure does not depend on R, i.e. p = constant, this equation can be integrated to obtain:

$$\left(\frac{dR}{dt}\right)^2 = \frac{8\pi p}{3m}\left(R_o^3 - R^3\right)$$

being R_o the radius of the core in the uncompressed state. Taking into account that m = (4/3)π R_o^3 ρ_o, with ρ_o the initial density of the sphere, the following relation between the implosion velocity u_p, the radius R and the applied pressure p, is derived:

$$u_p = \left\{\frac{2p}{\rho_o}\left[1-\left(\frac{R}{R_o}\right)^3\right]\right\}^{1/2} \qquad (1)$$

From Eq. (1) it is evident that, the higher is the pressure applied to the outer surface of the sphere, the higher are the particle velocities caused by the compression, the most significant is the value of α and, finally, also the nuclear explosive yield. It should be pointed out that Eq.(1) is valid in the hypothesis of constant pressure acting on the external surface of the sphere, i.e. the value of pressure does not change along the radius R. Exploratory calculations, performed by hydrodynamic codes, show that this approximation is reasonable in the case of limited penetration of the shock wave within the sphere (Kessler, 2008).

3.2 Equation of state in the compressed state

During the compression phase, a shock wave propagates inside the material with velocity D, while the particles move inward with velocity u_p. For many materials, a linear correlation between D and u_p has been found:

$$D = c_o + S\,u_p \qquad (2)$$

with c_o and S that depend on the material properties. In the case of plutonium metal, the parameters c_o and S assume the following values (Kessler, 2011): c_o = 2.51 x10^3 m/s, S = 1.3. Using the Rankine-Hugoniot equations and the Eq. (2), the following Hugoniot curve is obtained as function of parameters c_o and S (Kohn, 1969):

$$p_H = \frac{\rho_o \, c_o^{\,2} \mu \, (\mu + 1)}{[1 + \mu \, (1 - S)]^2} \qquad (3)$$

being p_H the pressure on the Hugoniot curve, ρ and ρ_o the density in the disturbed and undisturbed state, and $\mu = (\rho/\rho_o - 1)$. For a given material, the Hugoniot curve defines all the pressure-volume states obtainable through a shock transition. In the Figure 1, experimental data from Benedict et al., for the range up to 0.06 TPa, and from Kirzhnits and Hobel valid up to 0.1 TPa, are shown (Kessler, 2011). In the same figure, the Hugoniot curve from Eq.(3), with c_o = 2510 m/s and S =1.3, is also depicted for comparison purpose. It should be mentioned that pressures in the range of 0.06-0.08 TPa, produced by high-explosive, were available at the time of development of the first nuclear weapons.

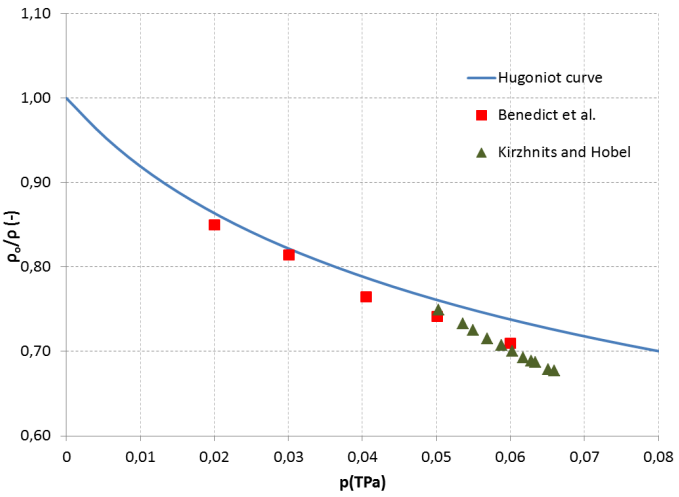

Figure 1 – Comparison between experimental data and the Hugoniot curve for plutonium metal.

4. Impact of spontaneous neutron fission emission on pre-detonation

An HNED with a strong spontaneous (fission) neutron emission would lead to early pre-detonation (also referred as pre-ignition) of the device, with a power excursion immediately upon prompt criticality. As consequence of this early pre-detonation, the shock wave is able to compress only the outer part of the fissile sphere; this fact limits the reactivity that can be inserted in the system and, then, the yield.

4.1 Probability of pre-detonation

Pre-detonation is a stochastic phenomenon and requires a treatment based on the probability theory. A theory of the spontaneous fission neutron emission has been developed by Hansen (Hansen, 1960) and it will be used, in the present work, to analyse the pre-detonation probability of an HNED based on the reactor-grade plutonium. In the hypothesis that a ramp of reactivity is inserted in the plutonium core, during the implosion phase, the cumulative probability $P(t_1)$ that a persistent fission chain occurs until the time t_1, i.e. integral probability to have pre-detonation between t= 0 (starting time of prompt criticality) and $t=t_1$, is given by the relation (Hansen, 1960):

$$P(t_1) = 1 - \exp\left(\frac{-\Delta k_{\max} \, S_M \, t_1^{\,2}}{\nu \, \Gamma_2 \, t_o}\right) \qquad (4)$$

where ν is the average number of prompt neutrons emitted per spontaneous fission, S_M is the total multiplied spontaneous fission neutrons rate, Δk_{max} is the maximum reactivity inserted in the fissile system during the time t_o, and Γ_2 is the Diven factor, equal to 0.8 for the Delta function distribution or "weak" neutron source (i.e. $S_M < 9\text{x}10^7$ n/s), and 1 for the Poisson distribution or "strong" neutron source. According to this theory, the average time $t_{1,ave}$ at which the pre-detonation occurs is given by:

$$t_{1,ave} = \left(\frac{\pi \nu \, \Gamma_2 \, t_o}{4 \, \Delta k_{\max} \, S_M}\right)^{0.5} \qquad (5)$$

This value can be assumed as the time at which the cumulative probability of pre-detonation is around 50%. As it can be seen from Eq. (4), the cumulative probability of pre-detonation $P(t_1)$ increases, for a given value of the reactivity Δk_{max}, with the increasing of time t_1 and with the neutrons rate S_M. The parameter S_M (n/s), in Eq. (4) and Eq. (5), takes into account two important aspects: firstly that the spontaneous neutrons that emerge homogeneously, from a sphere of fissile material, have a smaller importance than a point source in the center of the sphere, and secondly, the subcritical condition of the core; if FF is the form factor for the neutron flux distribution in a sphere of fissile material of mass M in subcritical condition (FF ≈ 0.81), S_M is related to the inherent fission neutron source $S_{inherent}$ (n/s) by the relation (Seifritz, 2009):

$$S_M = S_{inherent}\ 0.81\ \frac{1}{(1-k_{eff})} \qquad (6)$$

where $S_{inherent}$ is the total number of neutrons emitted inside the plutonium of mass M ($S_{inherent}$ = S x M, being S (n/kg/s) the spontaneous neutron fission source of Table 2) and the term $1/(1 - k_{eff})$ is the subcritical multiplication factor.

4.2 Probability of pre-detonation at different yields and minimum yield

A simplified approach to evaluate the cumulative probability of pre-detonation at different yields has been proposed by Mark et. al., in the hypothesis that $\Delta k_{max} = 1$ and that $e^Z = e^{45}$ are the fissions occurred before the maximum super-criticality is achieved (Mark, 1993). The results provided by Mark have been generalized by the author of this work, for any value of maximum reactivity Δk_{max} inserted in the system. In the hypothesis of ramp time function for the reactivity ($\Delta k = k - 1 = (t/t_o)\ \Delta k_{max}$), and using the Serber's Relation[d] (Serber, 1992), it is possible to derive an analytical relationship between the integral or cumulative probability for pre-detonation P and the nuclear explosive yield fraction Y/Y_o:

$$P\left(\frac{Y}{Y_o}<1\right)=1-\exp[S_M\ (Z\,l-\frac{\Delta k_{\max}}{2}\ x^{2/3}t_o)] \qquad (7)$$

being S_M, Z, l, Δk_{max}, t_o already defined and $x = Y/Y_o$, with Y the expected yield and Y_o the nominal (or design) yield. The previous relation evaluated for x=1, i.e. $Y = Y_o$, provides the cumulative probability to obtain an explosion with full yield Y_o. The theory also assures a minimum yield Y_{min} or "fizzle yield" given by:

$$\frac{Y_{\min}}{Y_o}=\left(\frac{2\,Z\,l}{\Delta k_{\max}\ t_o}\right)^{3/2} \qquad (8)$$

5. Explosive yield of HNEDs based on reactor-grade plutonium

This section is devoted to the evaluation of the explosive yield due to the detonation of the HNEDs based on the reactor-grade plutonium compositions of Table 1: RG(I), with an isotope composition from LWR spent fuel after an 60 GWd/t exposure, and RG(II) that represents a typical isotopic composition after three times recycling in a MOX-fueled LWR. Due to the high contents of ^{240}Pu isotope in both fuels, pre-detonation is taken into account.

[d] The so-called Serber Relation affirms that the energy yield of an exploding nuclear device is proportional to $(\Delta k_{max})^3$, and inversely proportional to l^2, being l the neutron life time : $Y \div (\Delta k_{max})^3 / l^2$. This is based on the assumption that the Rossi-α, α(t), and the outer radius of the plutonium sphere R(t), in the

5.1 Neutronic and physical parameters of cores

The analysis of the HNEDs will be done with reference at two bare spheres[e] having radii of 6.670×10^{-2} m for RG(I), and 7.185×10^{-2} m for RG(II). In the hypothesis of using plutonium metal, δ-phase (ρ_o = 15800 kg/m^3), the core masses are 19.64 kg and 24.55 kg, respectively. It is assumed that a uniform external pressure of 0.06 TPa acts on the outer surfaces of the cores, providing a compression ratio $c_r = \rho/\rho_o = 1.43$ (see Figure 2). The value of the 0.06 TPa has been chosen, being in the range of pressures considered in the design of first nuclear weapons of the implosion type (low technology). In the Table 3, the values of the isotopic mass M, the inherent fission neutron source $S_{inherent}$ and the total multiplied spontaneous fission neutrons rate S_M, for both the fuel compositions, are reported.

Table 3 - Reactor-grade plutonium compositions: mass and value of parameters S, $S_{inherent}$ and S_M.

Grade	Isotope	weight (%)	Mass (kg)	$S_{inherent}$ (n/s)	S_M (n/s)
RG(I)	^{238}Pu	3.8	0.746	1.940×10^6	9.410×10^7
	^{239}Pu	51.8	10.173	2.035×10^2	9.868×10^3
	^{240}Pu	23.1	4.537	4.128×10^6	2.002×10^8
	^{241}Pu	14.1	2.789	1.395×10^2	6.763×10^3
	^{242}Pu	71.	1.394	2.370×10^6	1.149×10^8
	Total	*100.0*	*19.639*	*8.439×10^6*	*4.093×10^8*
RG(II)	^{238}Pu	5.5	1.350	3.510×10^6	1.777×10^8
	^{239}Pu	34.1	8.371	1.674×10^2	8.476×10^3
	^{240}Pu	31.1	7.635	6.947×10^6	3.517×10^8
	^{241}Pu	10.6	2.602	1.301×10^2	6.587×10^3
	^{242}Pu	18.7	4.591	7.804×10^6	3.951×10^8
	Total	*100.0*	*24.549*	*1.826×10^7*	*9.245×10^8*

expansion phase of the nuclear explosion, are inversely proportional.

[e] The designs here considered do not include any reflector.

As reported in the Table 3, the reactor-grade plutonium RG(II) has a value of $S_{inherent}$ higher than that of the reactor-grade plutonium RG(I); assuming in both cases the same sub-critical level before compression (k_{eff} = 0.98), this difference also remains in terms of S_M. The value k_{eff} = 0.98 was chosen for two main reasons: i) safety reasons require a certain amount of sub-criticality, and ii) k_{eff} should not be too low if the compression must induce sufficient super-criticality. The calculation of the main neutronic parameters as the multiplication factor k_{eff}, neutron life time *l* and Rossi-α, has been performed by the MonteCarlo Reactor Physics code Serpent ver. 1.1.7, and using the JEFF-3.1 neutronic libraries (Leppanen, 2009); the results of these simulations are reported in the following Table 4.

Table 4 - Neutronic and physical parameters of two cores.

Grade	R (10^{-2} m)	c_r	k_{eff}	*l* (10^{-8} s)	α (10^6 s^{-1})
RG(I)	6.67	1.00	0.9833	0.371	-4.18(*)
	6.60	1.03	1.0004	0.368	-0.11(**)
	6.40	1.13	1.0544	0.359	14.05
	6.20	1.21	1.1117	0.350	29.73
	5.92	1.43	1.1975	0.336	55.27
RG(II)	7.18	1.0	0.9842	0.422	-3.42(*)
	7.12	1.03	0.9999	0.420	-0.08(**)
	6.90	1.13	1.0534	0.410	11.93
	6.60	1.29	1.1324	0.397	30.76
	6.38	1.43	1.1960	0.387	47.06

Note: (*) sub-critical state, (**) critical state

As reported in the previous Table 1, the reactor-grade plutonium RG(I) contains a higher isotopic concentration of ^{239}Pu and ^{241}Pu, in comparison with RG(II)[f] (66% against

[f] In the neutron energy range of 0.1-1MeV, most of the fissions are due to ^{239}Pu and ^{241}Pu, with a contribution of ^{240}Pu only above 1.0 MeV.

45%); this fact determines, for the same value of the compression ratios c_r, a higher value for α. In the Figure 2, the dependence of α and k_{eff} from the radius R, is reported for both fuels.

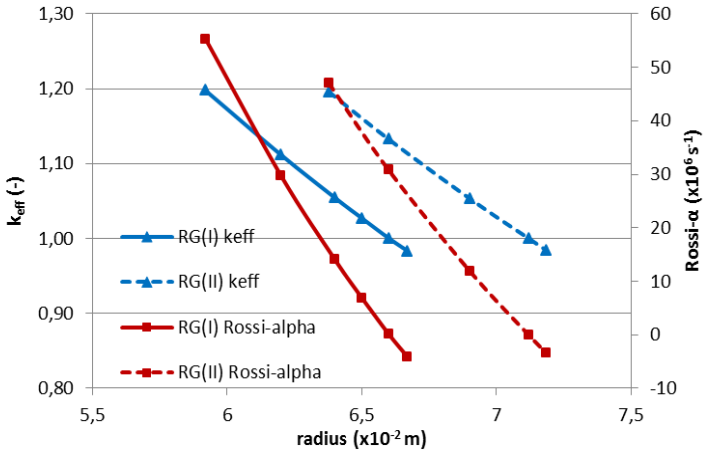

Figure 2 – k_{eff} and α as function of radius R for the two cores.

5.2 Time scale of the implosion processes

The implosion processes, described in the previous section, are characterized by a reduction in the radii of cores of less than one centimeter, and precisely: $\Delta R = 0.75 \times 10^{-2}$ m for RG(I) and $\Delta R = 0.81 \times 10^{-2}$ m for RG(II). Under these conditions, the calculation of the imploding velocity u_p is provided by the Eq. (1), so that the time scale of the implosion process can be derived. In a similar way, using the data of Table 4, it is possible to deduce the time-dependent behavior for α (sigmoidal curves), as shown in the Figure 3. In the same figure, the idealized reactivity ramps, starting from the prompt criticality (α=0), and to be used in the model of pre-detonation, are also shown for comparison purposes. Sigmoidal curves start from negative values (subcritical state) and reach the condition α=0 (prompt criticality) after 2.36×10^{-6} s for the reactor-grade plutonium RG(I), and 2.34×10^{-6} s for reactor-grade plutonium RG(II), from the beginning of compression; they, then, assume positive values (super-critical state) rising up to the final values $\alpha_o = 55.27 \times 10^6$ s^{-1} and $\alpha_o = 47.06 \times 10^6$ s^{-1} at the times 9.0×10^{-6} s and 9.74×10^{-6} s, respectively.

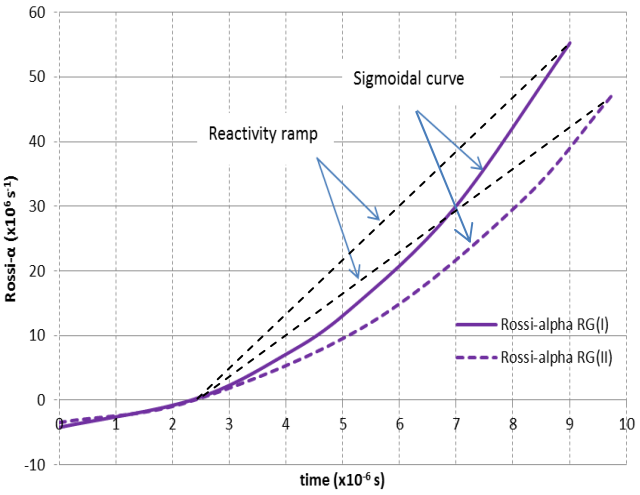

Figure 3 – Sigmoidal curves representing α as function of time.

The number of neutron generations Z, at the time of maximum compression, can be calculated for each core, by the following relation (Seifritz, 2009) :

$$Z = \ln\left(1 + \frac{M\ q_b\ \alpha_o}{P_o}\right) \qquad (9)$$

being M the core mass, q_b the specific internal energy at the boiling point (0.54×10^6 J/kg), α_o the maximum value reached by α, and P_o the initial power due to an external neutron source S_{eff} (with $S_{eff} > S_M$), switched on at the compressed state in order to ignite the chain reaction. As reported by Sandmeier (Sandmeier, 1972), Z is adjusted in such a way that from 37 to 40 neutron generations are produced at the time of maximum compression. Although this assumption looks arbitrary, it has no further consequence on the following sequence of events, being P_o negligible in comparison with the power released in the nuclear explosion. Assuming in the calculations the values

$P_o = 2.5x10^{-2}$ W for the reactor-grade plutonium RG(I), and $P_o = 5.6x10^{-2}$ W for reactor-grade plutonium RG(II), the number of neutron generations, at the time of maximum compression, are Z= 38 and Z = 37, respectively.

5.3 Calculation of the pre-detonation probability

According to Eq.(4), it can be evaluated the cumulative probability $P(t_1)$ that a persistent fission chain occurs until the time t_1, i.e. the integral probability to have pre-detonation between t= 0 (time of prompt criticality) and $t=t_1$. In addition, if Y_o is the full yield of the HNED, the cumulative probability of pre-detonation $P(Y/Y_o)$ at the fraction yield Y/Y_o, can be calculated by using Eq. (7). The main data considered for calculations are reported in the following Table 5 (ν =3.08, Γ_2 = 1 for both cores).

Table 5 - Data considered for the calculation of $P(t_1)$ and $P(Y/Y_o)$.

Grade	Δk_{max}	S_M (n/s)	t_o (s)	Z	l (10^{-8} s)
RG(I)	0.1975	$4.093x10^8$	$6.65x10^{-6}$	38	0.336
RG(II)	0.1960	$9.245x10^8$	$7.40x10^{-6}$	37	0.387

The cumulative probability of pre-detonation $P(t_1)$ as function of time is depicted in the Figure 4 (note that time t_1 is counted from the prompt criticality, i.e. $t_1 = 0$ at α=0). As it can be seen from the picture, the high isotopic content of ^{240}Pu and ^{242}Pu in the reactor-grade plutonium RG(II), in comparison with that in the RG(I), determines a higher value of $P(t_1)$ at any time; the average time $t_{1,ave}$ at which the pre-detonation occurs, i.e. the time at which the cumulative probability of pre-detonation is equal to 50%, is provided by Eq.(5): $t_{1,ave} = 4.460x10^{-7}$ s for RG(I) and $t_{1,ave} = 3.142x10^{-7}$ s for RG(II). Values of the function $P(Y/Y_o)$, for different fraction yields Y/Y_o, are reported in the following Table 6.

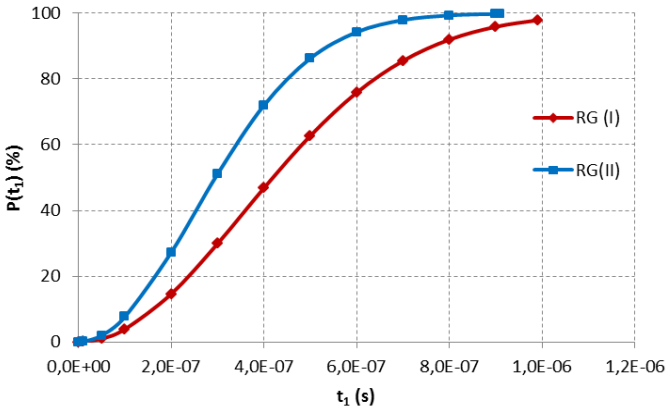

Figure 4 - Cumulative probability of pre-detonation $P(t_1)$ vs. time.

Table 6 - Cumulative probability of pre-detonation $P(Y/Y_o)$ for different fraction yields Y/Y_o.

RG(I)		RG(II)	
Y/Y_o	$P(Y/Y_o)$ (%)	Y/Y_o	$P(Y/Y_o)$ (%)
1.000	100.0	1.000	100.0
0.093	96.9	0.091	99.6
0.090	89.9	0.089	87.2
0.087	66.1	0.0885	65.1
0.086	49.2	0.088	42.4
0.085	23.7	0.0878	29.5
0.084	0.5	0.0875	0.8

In the following Figure 5, the cumulative probability $P(Y/Y_o)$ as function of the ratio Y/Y_o, is shown. As it can be deduced from the previous figure, both the HNEDs, based on the reactor-grade plutonium, lead to a quasi-deterministic yield equal to 8.4% of the full yield Y_o, for the first core with RG(I), and 8.7% of the full yield Y_o, for the second core with RG(II). These yield values are those that can be obtained by the use of Eq. (8), for the minimum yield Y_{min} or "fizzle yield".

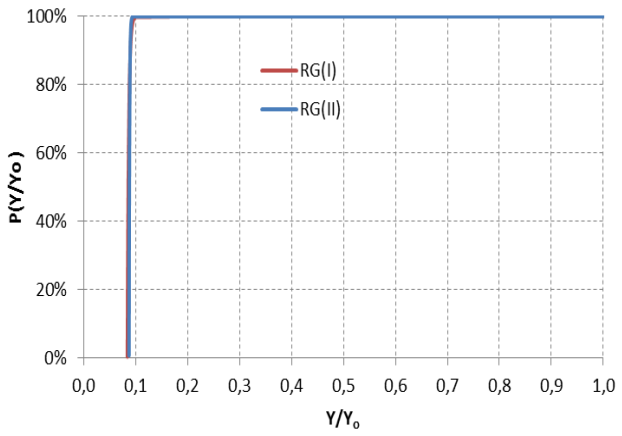

Figure 5 – Cumulative probability of pre-detonation $P(Y/Y_o)$ vs. the ratios Y/Y_o

5.4 Determination of the explosion yields

The determination of the design yield Y_o (in kiloton - TNT equivalent), associated to each HNED, can be done by using the analytical nuclear excursion model proposed by Seifritz (Seifritz, 2009), where the neutronic/hydrodynamic behaviour of the core is split in two parts: a time span up to the boiling point, without any relevant feedback mechanism, and a time span beyond the boiling point, where a strong feedback mechanism exists due to the expanding core volume. The results of the simulations give for two cores the following yields: Y_o =5.34 kT, for the core with isotopic composition RG(I), and Y_o = 5.68 kT for the core with isotopic composition RG(II); according to the quasi-deterministic value of the yields provided by the pre-detonation theory, the explosions will release: Y = 0.45 kT in the first case, and Y = 0.50 kT in the second case. These low values demonstrate as the pre-detonation phenomenon strongly affects the maximum yield achievable during the explosion; in fact, the pre-detonation induces the early disassembling of the device, before the maximum compression can be achieved, so limiting the maximum reactivity that can be inserted in fissile core. These conclusions are in agreement with those provided by the work of Kessler, where specific hydrodynamic calculations were employed to evaluate the explosion yield of a HNED, using plutonium from LWR spent fuel (Kessler, 2011). Although these yields are much smaller than the design yield (less than 10%), combined with thermal and prompt radiation effects, they would still constitute severely damaging explosions. No country has, so far, used reactor-grade plutonium for the construction of a nuclear explosive device. There are many difficulties that a proliferator would face to manufacture it: large critical mass, size and weight, small and unpredictable yield, high radiation dose. The degree to which these obstacles can be overcome, however, depends on the sophistication of the state or group attempting to produce a nuclear weapon. For that reason, any action enabling to reduce the proliferation risk associated to the use of the reactor-grade plutonium, as the possibility of denaturing it with isotopes that would make it more difficult to use as nuclear explosive (e.g. high ^{240}Pu concentration), as well as control and monitoring of plutonium inventory from recycling fuel, would be strongly supported and pursued (Polidoro, 2013).

6. Conclusions

The aim of this work was to evaluate, making use of available literature data and physical models, the potential nuclear explosive yield of an Hypothetical Nuclear Explosive Device of the implosion type, based on the reactor-grade plutonium. The study was focused on two nuclear explosive devices made up with plutonium compositions available after the reprocessing of traditional LWR fuels. The technology level here assumed, i.e. the capacity to reach high compression levels of the fissile material, is that assumed to be available at the time of the early development of nuclear weapons. The feature of the spontaneous fission neutron emission has been evaluated with a probabilistic approach, and the probability of pre-detonation was considered for both the cores. The results of analysis show that, due to the high isotopic concentration

of ^{240}Pu in the plutonium mixture, an early pre-detonation can occur with the effect to reduce the yield, i.e. to give a yield with low reliability. This fact, in addition to the heat generated by ^{238}Pu, and with the high radiation level that imposes careful management in the handling of fissile material, can poses serious problems to any proliferator nation or group, interested in manufacturing a weapon device based on such plutonium. However, due to the fact that the development of a low yield device based on such plutonium could not be excluded, the risk associated to the misuse of materials and technologies for the development of nuclear weapons based on this grade plutonium should be considered and assessed. In parallel, the development of nuclear fuels that, in any phase of the reactor cycle, present a plutonium isotopic composition not usable for weapon construction should be strongly encouraged.

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