

Computational model and physical and technical factors determining the plutonium proliferation resistance*

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Abstract

Since the closed nuclear fuel cycle suggests that plutonium is extracted from irradiated fuel and is recycled in nuclear reactors as part of the loaded fuel, proliferation resistance of fissile materials (plutonium) is becoming a problem of a practical significance. It is important to understand to what extent the physical and technical properties of fissile materials are capable to prevent these from being diverted to nonenergy uses. This paper considers the term "proliferation resistance" from a physical and technical point of view with no measures taken for the physical protection, accounting and control of nuclear materials. Thus, proliferation resistance of plutonium means that it is technically impossible to fabricate a nuclear explosive device (NED) of the implosion type due to the overheating of the device's components and the resultant NED failure.

The following conclusions have been made.

1. The assessment of the plutonium proliferation resistance is not justified where it relies on the analysis of an implosion-type NED excluding the use of modern heat-resistant and heat-conducting chemical explosives (CE) which are inaccessible.
2. Consideration of the asymptotic temperature profile in the NED components is not justified enough for the development of plutonium proliferation resistance recommendations.
3. No options enabling the slowdown of the NED warm-up process have been exhausted for analyzing the physical and technical factors that determine the proliferation resistance of plutonium.

General conclusion. The underlying rationale in a fundamental monograph by Dr. G. Kessler proved to be insufficiently valid, which has led to an unfounded inference as to the status of the plutonium proliferation resistance. The development of the procedures used and other factors taken into account are expected to increase the requirements to the content of the ²³⁸Pu isotope in plutonium for ensuring its proliferation resistance.

Keywords

Plutonium, plutonium-238; proliferation resistance; nuclear explosive device; explosive; cryogenic temperatures

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Introduction

It was reported in 1977 that the USA successfully tested in 1962 an NED based on reactor-grade plutonium (Gillette 1977). At the same time, in the late 1970s, strategies were analyzed within the IAEA framework for the nuclear fuel cycle closure (International Conference on Nuclear Power and its Fuel Cycle 1977), where an issue was raised concerning the proliferation resistance of reactor-grade plutonium. This problem was investigated by A. De Volpi (DeVolpi 1982) who came to the conclusion that “the best strategy to prevent the military use of plutonium is to introduce the ^{238}Pu isotope into it”. This plutonium isotope is an intense source of heat which can lead to an unacceptably high NED overheating (Carson 1993).

In the authors’ opinion, the problem of estimating quantitatively the fraction of ^{238}Pu required for the plutonium proliferation resistance was considered for the first time in (Heising-Goodman 1980). As plutonium is an intense source of spontaneous neutrons, and no efficient barrel-type NED can be built on its basis, (Heising-Goodman 1980) considers a model of an implosion-type NED in the form of a fissile material (plutonium) sphere surrounded by layers of tamper, the chemical explosive (EC) and the outer shell. A conclusion has been made that the introduction of plutonium, e.g., of at least 5% of ^{238}Pu , will lead to such high temperatures due to the heat release from the α -decay that the chemical explosive will melt down, which means that this implosion-type NED will be nonoperational (Heising-Goodman 1980).

Further evolution of the approach to estimating the ^{238}Pu fraction required for the plutonium proliferation resistance was presented in (Kessler 2007, Kessler and Chen 2008, Kessler 2008, Kessler 2011) authored by Dr. Gunther Kessler of Karlsruhe, Germany. As compared to his earlier study (Heising-Goodman 1980), Dr. G. Kessler’s works have the following specific features:

- a more justified NED structure is considered: the fissile material sphere radius is selected based on criticality calculations (several values of the fissile material sphere radius are considered in (Heising-Goodman 1980) with no reference to criticality);
- properties of various chemical explosives are analyzed to envisage the NED inoperability when any of the accessible chemical explosives is used in it;
- a circumstance is taken into account that the loss of the NED serviceability can be caused not only by melting but also by self-ignition and pyrolysis (decomposition with formation of gaseous components) of the chemical explosive.

Besides, a possibility is analyzed for the implosion-type NED warm-up process to be slowed down using such measures as prior cooling of the plutonium and the arrangement of a heat removing layer around the NED. It needs to be noted that such capabilities for the implosion-type

NED warm-up process slowdown were first considered by the authors in (Kulikov et al. 2009, Kulikov et al. 2010).

It is stated in the conclusion of monograph (Kessler 2011) that plutonium with the content of ^{238}Pu at a level of 11 to 13% can be regarded as proliferation-resistant, i.e., not fit for the fabrication of an implosion-type NED on its basis.

The major drawbacks of monograph (Kessler 2011) appear to be:

- the procedural aspects of the approach concerning the selection of the implosion-type NED structure and materials;
- consideration of the asymptotic temperature profile by the NED layers achieved upon an infinite warm-up time;
- no such capabilities for the implosion-type NED warm-up process slowdown are taken into account, as prior cooling of the neutron reflector (natural uranium), the tamper (aluminum) and the NED’s nonnuclear components, as well as the addition of heat-insulating layers to the NED structure, using which the temperature field can be redistributed such that the NED remains operational for as long as possible.

It can be however noted that Dr. G. Kessler’s monograph is practically the only exhaustive monograph in the 21st century devoted exactly to the physical and technical analysis of the reactor-grade plutonium proliferation resistance barriers. With a good academic standing enjoyed by its author, the monograph will be used by experts (and has already been used as practice shows) to develop a (U-Pu) cycle and to shape concepts of its resistance to uncontrolled proliferation of fissile materials.

The procedural aspects of Dr. G. Kessler’s approach are discussed in this paper, and opinions are given as to the imperfection of his analysis which led to the insufficiently valid inferences concerning the proliferation resistance of plutonium.

Model of a hypothetical implosion-type NED

The geometrical model of a hypothetical implosion-type NED is presented in Fig. 1 (Kessler 2011). The central plutonium charge is surrounded by spherical layers of natural uranium (neutron reflector), aluminum, the chemical explosive, and the outer shell.

When compressed, plutonium has its mass, being subcritical in the initial state, becoming more dense, supercritical and capable to produce an exponentially developing chain fission reaction. Due to a rapid change of neutron generations, this chain reaction has enough time to turn so intense (before the NED itself breaks down) that the total energy release becomes thousand times as great as the energy release from the chemical explosive.

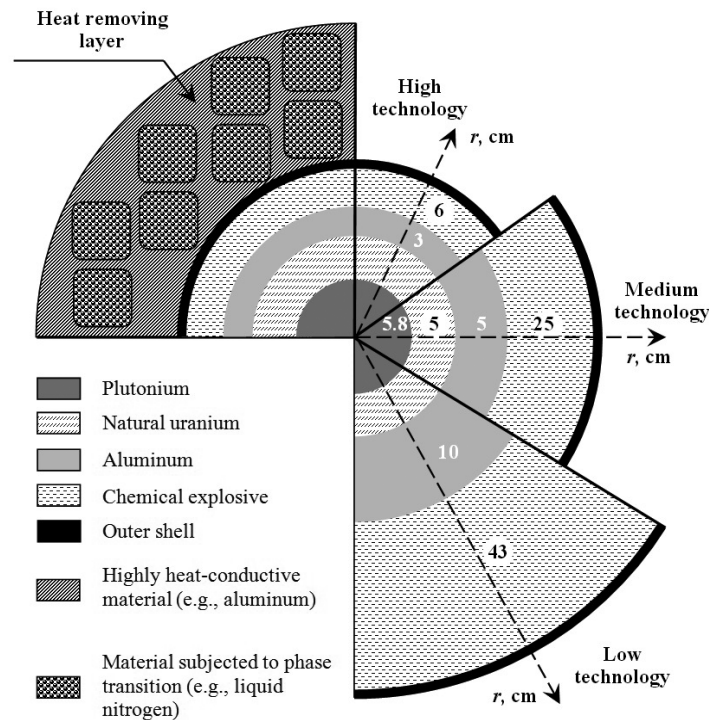


Figure 1. Geometrical model of a hypothetical implosion-type NED

Monograph (Kessler 2011) draws attention to the fact that the properties of any chemical explosive material are highly sensitive to temperature. In other words, its pyrolysis intensifies as the temperature increases, and it can melt down and self-ignite. In all of the above cases, the NED turns out to be nonoperational.

Dr.G. Kessler's philosophy to support plutonium proliferation resistance

Monograph (Kessler 2011) considers three technology evolution levels (see Fig. 1) of implosion-type NEDs:

- low (early NEDs of the mid-1940s);
- medium (NEDs of the 1950s);
- high (post-1960s NEDs).

These technology evolution levels reflect differences in the layout of the NED's nuclear components and the chemical explosive materials of different thermal stability used in the NED nonnuclear part. Thus, for example, a chemical explosive based on Composition B and baratol (melting temperature of 79 to 80 °C, intensive pyrolysis temperature of 180 to 200 °C, self-ignition temperature of 214 to 300 °C), which was used in the nonnuclear part of early NEDs, reflects the low level of technology (Composition B 2017, Baratol 2017).

This means that, in their nuclear part, the first NEDs used low-technology structural arrangements and designs of the mid-1940s. At the same time, only low-melting chemical explosive materials, also of the mid-1940s, are supposed to be used in the NED nonnuclear part. In the given case, this is a very important methodological and, at the same time, very strange assumption of the author made by him, as a matter of fact, in the second decade of the 21st century (i.e. 70 years after these early NEDs were built).

Indeed, the development and creation of compact NEDs are expected to reflect low- and high-technology levels, since such devices emerged, historically, much later than the initial models. At the present time, however, it can be hardly justified to say that heat-resistant chemical explosives (RVKh 9502, TATB) are also of a high-technology level which is as inaccessible as the creation of compact nuclear components for devices (Polymer-bonded explosive 2017, PBX 2017). Heat-resistant chemical explosives created in the 1970s are used, e.g., for blast hole drilling, are described in open literature (e.g., in (Manelis et al. 1996)), and do not contain any special materials. Chemical formulas of these heat-resistant explosives are well-known and information is also available on their fabrication technology. So the proposal by the author of (Kessler 2011) that the options reviewed should not include NEDs with low-tech nuclear components (from the mid-1940s), but with surrounding layers of heat-resistant explosives (developed in the 1960s and in the 1970s) in the nonnuclear part, cannot be looked upon as convincing.

A potential argument that these heat-resistant chemical explosive materials may be unavailable for use does not seem to be justified. In the long run, is not it impossible to buy components of a heat-resistant explosive and to produce the material of this explosive as such? Nuclear components as such (and their materials) are not required, as is known, for the development of the NED component that generates a spherically converging compression wave. It is exactly with no use of nuclear components that the implosion technology was developed in the mid-1940s.

The above argument is confirmed by not so old a precedent when the inaccessible centrifuge technology for the uranium isotope enrichment turned out to be accessible for the illegal network of the well-known Pakistani doctor Khan (Dr Khan: Nuclear Smuggler Broke the Silence 2016).

This made it possible for Pakistan to create its own A-bomb (Orlov 2002). Using his illegal network, Dr. Khan legally ordered and bought from companies in different countries of the world individual equipment parts and assemblies for gas centrifuges. This equipment was later mastered and used to build gas centrifuge cascades and to produce weapon-grade uranium for the Pakistani A-bomb.

It is therefore appropriate to consider models of devices with low-tech and high-tech nuclear components (as the “extreme” options) and to assume, for the generation of implosion, that the most heat-resistant chemical explosive, TATB, is always used (as that available at the present time) (Kessler 2007, Kessler and Chen 2008, Kessler 2008, Kessler 2011, Manelis et al. 1996).

Criteria determining nonserviceability of implosion-type NEDs

To incapacitate a NED, a heat source is proposed to be added to the fissile material to cause overheating of the NED components. The ^{238}Pu isotope (567 W/kg (Heising-Goodman 1980)) is considered as such heat source since, due to the heat from the α -decay, ^{238}Pu is exactly the most intense source of energy among the plutonium isotopes (Heising-Goodman 1980, Kessler 2007, Kessler and Chen 2008, Kessler 2008, Kessler 2011).

One needs to take into account all measures expected to prolong the period of time for which the NED will remain serviceable (we shall refer to this period as the NED “lifetime” and shall denote it as $\Delta\tau$). Therefore, the required level of the plutonium proliferation resistance (through the addition of the ^{238}Pu isotope) is justified based on analyzing the warm-up of differently structured implosion-type NEDs in various heat removal conditions and with the selection of the option that will ensure the device serviceability for as long time as possible. The fraction of the ^{238}Pu isotope, with which the NED will be

operational for quite a short time even in this case, can be regarded sufficient for considering the plutonium with such composition proliferation-resistant.

The following measures can be taken to prolong the NED lifetime:

- prior cooling of the NED components;
- arrangement of a heat removing layer around the NED;
- addition of a heat-insulating layer to the NED structure to enable such temperature field redistribution as will ensure the NED serviceability for the longest time possible.

We shall assume a multilayer implosion-type NED to have a spherically symmetrical geometry. We shall also proceed from the fact that the source of heat from the α -decay is spherically symmetrical, as the boundary conditions on the outer surface are also spherically symmetrical and the initial temperature distribution is described as well by a spherically symmetrical function.

In this case, the temperature distribution by the NED layers can be found from the solution of a non-steady-state heat conductivity equation:

$$\text{div} [\lambda(r, T) \times \text{grad } T(r, \tau)] + q_r(r) = c_r(r, T) \times \delta T(r, \tau) / \delta \tau,$$

where $\lambda(r, T)$, $c_r(r, T)$ is the material’s heat conductivity and specific heat capacity depending on the temperature $T(r, \tau)$; $T(r, \tau)$ is the temperature at the point r at the time τ ; and $q_r(r)$ is the heat source intensity.

The least heat-resistant implosion-type NED component is chemical explosive. We shall note that, apart from melting and self-ignition, there is one more process capable to lead to the loss of the chemical explosive serviceability, known as pyrolysis.

According to (Manelis et al. 1996), decomposition of 2 % of its molecules can be taken as the criterion of the chemical explosive stability. The decomposition rate can be estimated using the Arrhenius equation (Stiller 1989). Knowing the decomposition rate, one can easily determine the share of the chemical explosive (ε) that has decomposed by the given time.

The overwhelming majority of chemical explosives melt at a lower temperature than their self-ignition temperature, that is, melting is a more stringent criterion. And, depending on the considered chemical explosive, its melting can take place both earlier and later than 2 % of its molecules decompose. In particular, melting is observed earlier than 2 % of the molecules decompose in the event of TNT, while decomposition of 2 % of the molecules occurs earlier than melting for TATB.

It can be therefore concluded that, depending on the chemical explosive used, the most stringent criterion that defines the limited serviceability of implosion-type NEDs can be both melting of the chemical explosive and decomposition of 2 % of the chemical explosive’s molecules.

Rationale for the need to consider the implosion-type NED non-steady–state warm-up process

The NED cooling in this paper means that the NED components are cooled prior to the final assembly. And plutonium and uranium are cooled to a small extent (or not cooled at all), while aluminum and the chemical explosive (the materials with the highest heat-storing capacity) are cooled to lower temperatures. Such prior cooling is considered based on the assumptions that the NED remains serviceable for as long as possible with the plutonium temperature to be, however, in excess of 198 K throughout the warm-up period. The thing is that plutonium stabilized in the δ -phase transforms into the α' -phase at lower temperatures, which is accompanied by an abrupt volume change; the opposite transformation into the δ -phase is not fully reversible (Plutonium. Fundamental problems 2003). Eventually, such phase transformations may lead to the partial or complete loss of the NED serviceability.

The cooling is stopped immediately prior to the final assembly stage, after which the NED is assembled in quite a short time and transported further in the assembled form, now with no cooling equipment, but thermally insulated from the environment to keep the device cooled for as long time as possible.

It is important to note that the NED's external components (all of the device's components except plutonium and uranium) can be assembled at the indoor temperature with no time limits. The assembled external part of the NED is then pre-cooled. Following the pre-cooling, only the final NED assembly is required which includes the plutonium and uranium insertion into the semi-spheres of the device's external part and arrangement of the heat-insulating layer around the NED.

For this study, the NED serviceability can be lost only after the NED components are withdrawn from the cooler.

In this connection, the following criterion for the device nonserviceability can be proposed: an implosion-type NED is not fit for practical use if it loses serviceability in a shorter time than required for its final assembly, transportation and use.

Therefore, this procedure, based on the time factor, requires consideration of the implosion-type NED non-steady-state warm-up process so that to compare the time for the NED components to reach critical temperatures with the time needed for the device final assembly, transportation and use.

Conclusion

It can be concluded that there is a doubtful point in the fundamental monograph by Dr. G. Kessler (Kessler 2011) that the barrier defining the plutonium proliferation resistance was interconnected, in principle, directly to the author-suggested inaccessibility barrier of a standard heat-resistant explosive. The insufficiently valid underlying rationale led to an unfounded inference concerning the status of the plutonium proliferation resistance.

Besides, consideration of the asymptotic temperature profile in the NED components is not justified enough for the development of the plutonium proliferation resistance recommendations since the device final assembly, transportation and application stage requires a certain finite time period.

Apart from this, no additional capabilities for slowing down the NED warm-up process have been exhausted.

Therefore, the authors have developed a computational and theoretical model for assessing the plutonium proliferation resistance which lacks the drawbacks of earlier studies on the subject. The developed model is based on the justified NED structure, considers the non-steady-state warm-up process, and takes into account additional measures for the warm-up process slowdown.

References

- Baratol (2017) Baratol. <https://en.wikipedia.org/wiki/Baratol> [accessed Apr. 11 2017]
- Carson Mark J (1993) Explosive Properties of Reactor-Grade Plutonium. *Science & Global Security* 4: 11–128.
- Composition B (2017) Composition B. https://en.wikipedia.org/wiki/Composition_B [accessed Apr. 11 2017]
- DeVolpi A (1982) Denaturing Fissile Materials. *Progress in Nuclear Energy* 10(2): 161–220. [https://doi.org/10.1016/0149-1970\(82\)90022-1](https://doi.org/10.1016/0149-1970(82)90022-1)
- Dr Khan (2016) Dr Khan: Nuclear Smuggler Broke the Silence. <http://www.atominfo.ru/news/air7423.htm> [accessed Oct. 22 2016, in Russian]
- Gillette Robert (1977) Impure Plutonium Used in '62 A-Test. *Los Angeles Times*, September 16, Part 1: 3.
- Heising-Goodman CD (1980) An Evaluation of the Plutonium Denaturing Concept as an Effective Safeguards Method. *Nuclear Technology*, 50: 242–251. <https://doi.org/10.13182/NT80-A32527>
- International Conference on Nuclear Power and its Fuel Cycle (1977) Vol. 2. The nuclear fuel cycle, part 1. Salzburg, Austria, 2–13 May.
- Kessler G (2008) Proliferation Resistance of Americium Originating from Spent Irradiated Reactor Fuel of Pressurized Water Reactors, Fast Reactors and Accelerator-Driven Systems with Different Fuel Cycle Options. *Nuclear Science & Engineering* 159: 56–82. <https://doi.org/10.13182/NSE159-56>
- Kessler G (2011) Proliferation-Proof Uranium. Plutonium Fuel Cycles. Safeguards and Non-Proliferation. Germany, KIT Scientific Publishing.

- Kessler G (2007) Plutonium Denaturing by ^{238}Pu . *Nuclear Science and Engineering*, 155: 53–73. <https://doi.org/10.13182/NSE07-A2644>
- Kessler G, Chen X-N (2008) Thermal Analysis of Hypothetical Nuclear Explosive Devices Containing Reactor-grade Plutonium with Higher Content of Pu-238. *International Workshop on Non-proliferation of Nuclear Materials*. Obninsk, Russia, 29 Sep. – 3 Oct.
- Kulikov E, Shmelev A, Apse V, Kulikov G (2009) Mathematical models for quantitative evaluation of fissionable materials proliferation protection. *American Nuclear Society. IV-th Topical Meeting on Advances in Nuclear Fuel Management (ANFM IV)*. Hilton Head Island, South Carolina, USA, April 12–15.
- Kulikov YeG, Shmelev AN, Apse VA, Kulikov GG (2010) Calculational models for quantitative evaluation of proliferation protection for fissionable materials. *Izvestiya vuzov. Yadernaya energetika* 2010(2): 184–195. [in Russian]
- Manelis GB, Nazin GM, Rubtsov YuI, Strunin VA (1996) *Thermal decomposition and burning of explosives and powders*. Moscow. Nauka Publ., 224 pp. [in Russian]
- Nadykto BA, Timofeyeva LF (2003) *Plutonium. Fundamental problems Vol. 2*. Ed. by Sarov, RFNC-VNIIEF Publ. [in Russian]
- Orlov VA (2002) *Nuclear non-proliferation. Vol. 1*. Moscow. PIR-Center Publ., 528 pp. [in Russian]
- PBX (2017) PBX. <https://en.wikipedia.org/wiki/PBX> [accessed Apr. 11 2017]
- Polymer-bonded explosive (2017) Polymer-bonded explosive. https://en.wikipedia.org/wiki/Polymer-bonded_explosive [accessed Apr. 11 2017]
- Stiller W (1989) *Arrhenius Equation and Non-Equilibrium Kinetics. 100 Years of the Arrhenius Equation*. Leipzig, BSB B. G. Teubner Verlagsgesellschaft.