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February 27, 2017

The Myth of "Denatured" Plutonium
Reactor-Grade Plutonium and Nuclear Weapons

Part Five: Radiation and Critical Mass

This paper is the fifth in a series to comprehensively examine the nuclear weapon dangers posed by reactor-grade plutonium. The first paper described some of the basic properties of plutonium, how it is classified into different grades, the variation in reactor fuel burnup and how plutonium's properties can vary depending on the initial fuel enrichment and burnup of the reactor fuel that produces the plutonium.[2] The second paper provided a short history of views regarding the nuclear weapon dangers of reactor-grade plutonium and discussed how the nuclear industry's desire to recycle plutonium has led it to downplay its dangers.[3] The third paper showed that the problem of the predetonation of an unboosted implosion fission weapon is not an impediment to the use of reactor-grade plutonium to produce nuclear weapons.[4] The fourth paper demonstrated that the increased heat content of reactor-grade plutonium produced in Light Water Reactors (LWRs) does not prevent such material from being used to produce nuclear weapons.[5]

Though predetonation and heat are the two main reasons often cited to support the mistaken notion that plutonium can be denatured, there are two other factors that are sometimes cited as well. These are the increased radiation of reactor-grade plutonium and its increased critical mass. In this paper I will demonstrate that neither of these factors poses a serious problem for the use of reactor-grade plutonium to produce nuclear weapons.

Radiation from Plutonium

All of the isotopes of plutonium are radioactive and therefore give off ionizing radiation as they decay. Of the five main isotopes (Pu 238, Pu 239, Pu 240, Pu 241 and Pu 242) that comprise reactor-grade plutonium, four (Pu 238, Pu 239, Pu 240 and Pu 242) decay by emitting an alpha particle. Alpha particles are quite short-ranged and easily blocked by even a piece of paper. Therefore as long as precautions are taken to prevent the plutonium from being inhaled or

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[1] This paper is the product of the author's personal research and the analysis and views contained in it are solely his responsibility. Though the author is also a part-time adjunct staff member at the RAND Corporation and a faculty member of the Pardee RAND Graduate School, this paper is not related to any RAND project or the Pardee RAND Graduate School and therefore these organizations should not be mentioned in relation to this paper. I can be reached at GregJones@proliferationmatters.com

[2] Gregory S. Jones, "The Myth of "Denatured" Plutonium, Reactor-Grade Plutonium and Nuclear Weapons, Part One: Introduction, Plutonium Basics, Definitions of Grades of Plutonium, Variation in Fuel Burnup, and the Properties of Plutonium Produced in Different Reactor Fuels," July 26, 2016. Link

[3] Gregory S. Jones, "The Myth of "Denatured" Plutonium, Reactor-Grade Plutonium and Nuclear Weapons, Part Two: Short History of Reactor-Grade Plutonium and Plutonium Recycle: Why Does the Nuclear Industry Downplay the Danger of Reactor-Grade Plutonium, September 1, 2016. Link

[4] Gregory S. Jones, "The Myth of "Denatured" Plutonium, Reactor-Grade Plutonium and Nuclear Weapons, Part Three: Predetonation," October 25, 2016. Link

[5] Gregory S. Jones, "The Myth of "Denatured" Plutonium, Reactor-Grade Plutonium and Nuclear Weapons, Part Four: Heat," December 15, 2016. Link

ingested, the alpha particles pose no radiation hazard. However, after the alpha particle is emitted, the resulting nucleus is sometimes left in an excited state which leads to the emission of a gamma ray or an x-ray. These radiations are far more penetrating than are alpha particles and can be a hazard to personnel though the gamma rays from the decay of these plutonium isotopes are generally low energy. Note that the daughter products of these four isotopes (U 234, U 235, U 236 and U 238 respectively) are all long-lived alpha emitters which do not contribute any significant radiation.

The decay of Pu 241 is different. Pu 241 decays by emitting a beta particle with a half-life of 14.4 years. Beta particles are somewhat more penetrating than are alpha particles but they are still only a hazard if the Pu 241 is ingested, inhaled or comes into direct contact with the skin. The beta decay of Pu 241 does not emit any gamma rays or x-rays. However, Pu 241's decay product, Am 241, with a 433 year half-life, emits significant amounts of low energy gamma radiation during its decay. Furthermore Pu 241 also rarely (half-life of about 600,000 years) emits alpha particles producing the decay product U 237 which has a half-life of only 6.75 days. The decay of U 237 produces relatively powerful but still low energy gamma rays.

Therefore almost all of the gamma radiation emitted by the decay Pu 241 is from its decay products and not the Pu 241 itself. When plutonium has first been separated from spent fuel, the plutonium is pure and the gamma radiation resulting from the decay of Pu 241 is very low. Quickly the amount of U 237 builds up in the plutonium and in only 6.75 days it is already half its equilibrium value and will reach its equilibrium value in about 50 days.[6] At the same time the Pu 241 is also decaying into Am 241 and due to Am 241's long half-life, it quantity increases steadily for many years. Initially the gamma radiation contribution from U 237 is dominant but in time the Am 241 overtakes it. In a situation where there is no shielding, the contribution from Am 241 becomes more important in just three months but in situations where there is significant shielding, it can take years.

Reactor-grade plutonium gives off significantly more gamma radiation than does weapon-grade plutonium due to increased amounts of Pu 241 and Pu 238. At the time of chemical separation, reactor-grade plutonium gives off about four times as much gamma radiation as does weapon-grade plutonium. Over time, as the Pu 241 decay products accumulate, this ratio increases. At 50 days after separation the ratio is about a factor of 12 and one year after separation the ratio is about a factor of 14.[7]

This higher dose is not a significant impediment to using reactor-grade plutonium in nuclear weapons since the dose can be greatly reduced by a combination of shielding and keeping some distance away from the radiation source. Heavy elements used as shielding such as lead or

[6] The quantity of U 237 will be at equilibrium when the amount of U 237 produced by the decay of Pu 241 equals the amount of U 237 that decays away. The fraction of the equilibrium value attained is dependent on the half-life of U 237 and is found by the formula $1 - e^{-\lambda t}$ where λ is the ln 2 divided by the half-life.

[7] For this calculation, weapon-grade plutonium has the composition of 93.4% Pu 239, 6.0% Pu 240 and 0.6% Pu 241. Reactor-grade plutonium has the composition of 2.6 % Pu 238, 54.3% Pu 239, 25.8% Pu 240, 9.7% Pu 241 and 7.6% Pu 242. These calculations used equation 25.6 in H. V. Larson, "Factors in Controlling Personnel Exposure to Radiations from External Sources," *Plutonium Handbook*, Volume II, O. J. Wick ed., United States Atomic Energy Commission, 1967, p 851.

uranium are very effective at stopping the low energy gamma radiation from plutonium, Am 241, and U 237.

Keeping some distance away from a radiation source can also be very effective. Simple geometry demonstrates that the intensity of any radiation source declines with the inverse square of the distance from the source. This rule is not restricted to sources of ionizing radiation (gamma rays, x-rays and neutrons) but any source of radiation such as light from a light bulb. From our own experience we know that the light grows dim as we move away from a light bulb. The radiation level at the surface of a six kilogram plutonium sphere is reduced by a factor of 500 at one meter away.

The gamma radiation from plutonium spheres used as the cores of nuclear weapons can be very effectively shielded by covering them with just one centimeter of natural uranium. This would amount to about 6 kilograms of uranium and it could function as part of the weapon's tamper. Even for the relatively energetic gamma rays from U 237, the uranium layer would reduce the exposure by a factor of about 20,000. Many of the weaker gamma rays from plutonium and Am 241 would be stopped completely.

The dose from neutrons produced by spontaneous fission in metallic plutonium is less than that from gamma rays but neutrons are harder to shield against and in some circumstances can be more important. However, for safety and security reasons early nuclear powers (such as the U.S. in the 1940s and 1950s and Pakistan today) keep their cores stored away from the rest of the weapon. The stored cores can be heavily shielded and in peacetime personnel would have no reason to be near them. If the cores are removed from storage for combat use, the amount of time that personnel would have to be within a meter or less from the core would only be a matter of hours and would not result in a serious exposure.

For a six kilogram sphere of reactor-grade plutonium, the surface dose rate from neutrons would be about 1.2 rad/hr. The addition of 1 centimeter of natural uranium to the plutonium sphere would do little to reduce this dose rate. However, at 1 meter away from the sphere, the dose rate would only be 2.5 mrad/hr. Even at this close distance it would take 2,000 hours (a quarter of a year) to accumulate the 5 rem that is the U.S. standard for annual worker exposure to radiation.[8]

Processing of the reactor-grade plutonium from plutonium oxide into plutonium metal weapon cores could lead to increased exposure but it is not clear that the total exposure would be that significant. Countries such as Pakistan, India and North Korea are only adding about 5 new nuclear weapons to their arsenals each year. Therefore the amount of time workers would spend processing the reactor-grade plutonium for this small number of weapons would not be that great. In addition, the greatest exposure would be to workers hands. The U.S. standard for annual worker radiation exposure to the extremities is 50 rem.

Further some of the processing of even weapon-grade plutonium (in particular when it is a fluoride) requires remote handling. This raises the issue of what other plutonium handling operations could be handled remotely. When the U.S. developed its nuclear weapon production capacity in the 1940s and 1950s, there was little choice and most operations had to be performed

[8] For our purposes, the radiation units rad and rem are equivalent.

hands-on. With today's computer controlled machines, it could be possible for a new nuclear power to carry out many more operations remotely, making the increased radiation dose from reactor-grade plutonium largely irrelevant. Processing the reactor-grade plutonium just after it has been chemically separated would be another method for reducing worker radiation exposure.

Another far cruder alternative would be for the worker exposure levels to be higher than what the U.S. would consider acceptable. For example, in the first few years of the Soviet nuclear weapons program, workers were exposed to an average of 25 to 30 rem per year, which is five to six times the current U.S. standard for maximum worker exposure.[9]

Some have asked that if reactor-grade plutonium can be used to produce nuclear weapons, why has the U.S. not used it. There are multiple reasons. First, it should be remembered that the U.S. infrastructure to produce weapon-grade plutonium was built in the 1940s and 50s when there was no reactor-grade plutonium. By the mid-1960s when reactor-grade plutonium first started to become available, U.S. production of weapon-grade plutonium was sharply declining as various plutonium production reactors were being shut down. U.S. production of weapon-grade plutonium ended for the most part by 1971. By that time the U.S. had plenty of weapon-grade plutonium and had no need to supplement it with reactor-grade plutonium.

Second, the increased radiation of reactor-grade plutonium is a major impediment to its use in U.S. nuclear weapons. U.S. radiation standards require not only that worker exposure be below specific limits (for example 5 rem for annual whole body exposure) but also that the ALARA principle be applied. ALARA stands for "As Low As is Reasonably Achievable," which means making every reasonable effort to maintain exposures as far below the dose limits as practical.[10] For the U.S. to change over to the use of reactor-grade plutonium while continuing to use the current weapon manufacturing facilities would result in increased worker radiation exposure which would be inconsistent with ALARA. U.S. weapon manufacturing facilities would need to be completely rebuilt in order not to increase worker radiation exposure. New nuclear proliferants are not bound by ALARA.

Third, changing from weapon-grade plutonium to reactor-grade plutonium in U.S. nuclear weapons would probably require that the weapon be recertified by conducting nuclear tests. However, the U.S. has had a nuclear test moratorium since 1992 which would prevent any such recertification. Since the U.S. has surplus weapon-grade plutonium, this is not an issue but for countries without large stockpiles of weapon-grade plutonium, reactor-grade plutonium could be an attractive alternative.

Radiation from other Nuclear Weapon Fissile Materials

Reactor-grade plutonium is not the only fissile material that can be used to produce nuclear weapons that emits significant amounts of gamma radiation. For U 233 the problem of gamma radiation can be substantially worse. U 233 is produced by irradiating thorium in nuclear

[9] TV Azizova, ES Grigorieva, MV Bannikova, and MB Moseeva, "Circulatory diseases in the cohort of Mayak PA workers occupationally exposed to radiation," *Joint RERF-ICRP Workshop on Health Risk of Radiation and the System of Radiological Protection*, Tokyo, Japan, October 9, 2016.
[10] 10 CFR 20.1003.

reactors. The resulting uranium is about 98% U-233 and about 1% each of U 234 and U 238. However, the production of U 233 also produces small quantities of U 232. Initial U.S. efforts to produce U 233 resulted in a U 232 content of over 100 ppm[11] but techniques were developed that resulted in U 233 containing only about 5 to 10 ppm U 232.[12]

U 232 has a half-life of 68.9 years and alpha decays into thorium 228 which has a half-life of 1.9 years. Th 228 then rapidly undergoes five more decay steps before ending as a stable lead isotope. One of Th 228 decay products (thallium 208) emits a powerful gamma ray that is difficult to shield against.[13] When U 233 is first separated from thorium, its radiation is low since there is no Th 228. The radiation buildup follows a pattern similar to that of U 237 from Pu 241 except since the half-life of Th 228 is 1.9 years as opposed to 6.75 days for U 237, it occurs much more slowly. U 233 containing just 5 to 10 ppm U 232 can be processed by hand in glove boxes by performing the operations soon after the U 233 is separated from thorium.

Handling fabricated U 233 weapon cores would be more difficult. One year after separation, U 233 containing just 5 ppm of U 232 emits seven times as much gamma radiation as does reactor-grade plutonium.[14] U 233 containing 100 ppm U 232 emits 150 times as much gamma radiation as does reactor-grade plutonium. A centimeter coating of natural uranium would only reduce this radiation by about a factor of two.

Yet even U 233 containing 100 ppm U 232 is usable in a nuclear weapon. "This emission [the gamma ray from thallium 208] produces a radiation field that requires much of the material to be stored inside shielded vaults. The radiation is sufficient to create major handling complications, but is not sufficient to prevent its use as a weapons-usable material."[15] Presumably the U 233 would be managed by keeping personnel away from the cores most of the time. The U.S. and the Soviet Union are each known to have conducted at least one nuclear test using U 233.[16]

Neptunium is now known to be a nuclear material that can also be used as the core of a nuclear weapon. Its gamma ray emissions are significantly less than those from U 233 but are similar to those from reactor-grade plutonium. Neptunium is produced by the irradiation of U 235 and in LWRs is produced at a rate about one-tenth that of plutonium. The isotope Np 237 is produced by this process and has a half-life of 2.1 million years. It decays into protactinium 233 by emitting an alpha particle. Pa 233 has a 27 day half-life and decays by emitting a beta particle and gamma rays. This process is similar to the production of U 237 by Pu 241 and the gamma rays from Pa 233 have a similar energy to those of U 237.

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[11] C.W. Forsberg, *et. al.*, "Disposition Options for Uranium-233," ORNL/TM-13553. Oak Ridge National Laboratory, June 1, 1999, Table 2.2, p. 12.

[12] J. M. Boswell *et. al.*, "Production of ^{233}U with Low ^{232}U Content," *Thorium Fuel Cycle*, Proceedings of Second International Thorium Fuel Cycle Symposium, Gatlinburg, Tennessee, May 3-6, 1966, U.S. Atomic Energy Commission, February 1968, pp. 745-763.

[13] P.J. Bereolos *et. al.*, "Strategy for the Future Use and Disposition of Uranium-233: Technical Information," ORNL/TM-13552, April 1998, Figure 2.2, p .5.

[14] Jungmin Kang and Frank N. von Hippel, "U-232 and the Proliferation Resistance of U-233 in Spent Fuel," *Science and Global Security*, Volume 9, 2001, Table 2, p. 10.

[15] Dean R. Tousley, Charles W. Forsberg and Alan M. Krichinsky, "Disposition of Uranium-233," ORNL/CP-95149, October 16, 1997.

[16] David Holloway, "Research Note: Soviet Thermonuclear Development," *International Security*, Winter 1979/80, Vol. 4, No. 3, p. 195.

The half-life of Np 237 is about three and one-half times longer than that of the alpha decay half-life of Pu 241 but neptunium is 100% Np 237 whereas reactor-grade plutonium is only about 10% Pu 241. As a result the gamma radiation would be about three times stronger from neptunium than from reactor-grade plutonium. The four times longer half-life of Pa 233 compared to U 237 means that there would be more time to process the neptunium before the gamma emissions reached their full intensity.

When handling a six kilogram sphere of neptunium to determine its critical mass, it was necessary to coat the neptunium with 0.28 cm of tungsten and 0.39 cm of nickel to reduce the gamma ray contact dose of the sphere to 300 mR/hr.[17] I calculate that even this small amount of shielding was able to reduce the gamma dose by about a factor of nine.

That neptunium and especially U 233 can be used to produce nuclear weapons is a clear indication that the increased radiation from reactor-grade plutonium will not prevent its use in a nuclear weapon.

Critical Mass

The critical mass of reactor-grade plutonium is larger than that of weapon-grade plutonium but it was authoritatively shown by Robert W. Selden of Lawrence Livermore Laboratory as long ago as 1976 that the critical mass of reactor-grade plutonium is significantly less than that of highly enriched uranium (HEU) and therefore can be readily used to produce nuclear weapons.[18] However, one still finds statements that do not accurately reflect the relative critical masses of these different types of plutonium. For example it has been claimed that while a nuclear weapon would require only three kilograms of weapon-grade plutonium, using reactor-grade plutonium would require eight kilograms and using plutonium recovered from mixed oxide fuel (MOX—fuel that was initially plutonium and uranium oxides) would require over 20 kilograms.[19] Therefore it is useful to review this issue.

Part of the confusion concerning the critical mass of reactor-grade plutonium is that the isotopes Pu 238, Pu 240 and Pu 242 are not readily fissioned by thermal neutrons. The isotopes Th 232 and U 238 are also not readily fissioned by thermal neutrons and these latter isotopes cannot sustain the fast neutron chain reaction needed to produce a nuclear explosion. It was initially assumed that these three plutonium isotopes could not sustain a fast chain reaction as well and their presence in reactor-grade plutonium would act as inert diluents. As long ago as 1969 it was known that this was not always the case and that some isotopes that cannot sustain a thermal chain reaction can sustain the fast neutron chain reaction needed to produce a nuclear explosion.[20] Selden showed that all three of these even-numbered plutonium isotopes can sustain a fast neutron chain reaction. More modern information has demonstrated that all the isotopes of neptunium, plutonium, americium and curium, which have half-lives of greater than ten years,

[17] Rene G. Sanchez, *et.al.*, "Criticality of a ^{237}Np Sphere," Los Alamos National Laboratory, 2003.
[18] Robert W. Selden, "Reactor Plutonium and Nuclear Explosives," Lawrence Livermore National Laboratory, 1976.
[19] Bruno Pellaud, "Proliferation aspects of plutonium recycling," *Journal of the Institute of Nuclear Material Management*, Fall 2002, p. 4.
[20] S. R. Bierman and E. D. Clayton, "Criticality of Transuranium Actinides-Undermoderated Systems," *American Nuclear Society Transactions*, Vol. 12, 1969. At that time data was only available for Pu 238 and Cm 244.

can sustain a fast neutron chain reaction. Indeed, of all the long-lived actinide isotopes only Ac 227, Th 230, Th 232, U 236 and U 238 cannot sustain a fast neutron chain reaction.[21]

The unreflected[22] critical masses of various nuclear materials relevant to the production of nuclear weapons are shown in Table 1.[23] Not all critical masses have been determined by direct measurement but are instead based on nuclear calculations. Therefore the estimated critical masses are a range, since different computer codes give different results. As can be seen, Pu 238 and Pu 241 have critical masses very similar to that of Pu 239. Pu 240's critical mass is somewhat larger but still less than that of HEU. Only Pu 242's critical mass is larger than that of HEU but even in plutonium produced in high burnup LWR fuel, this isotope is less than eight percent of the total plutonium and will not result in a great increase in the critical mass. Direct measurement of the critical mass of relatively low-burnup reactor-grade plutonium compared to weapon-grade plutonium shows only a 14% increase. For high-burnup reactor-grade plutonium the critical mass increase would be no more than about 30% which it less than half that of HEU. Such material could easily be used to produce nuclear weapons.

The critical mass of U 233 is similar to that of weapon-grade plutonium. The critical mass of neptunium is similar to that of HEU, which explains why this material is now considered capable of being used to produce a nuclear weapon. There is still a significant range of estimates for the critical mass of Am 241 and therefore its usefulness as a nuclear weapon material must be considered somewhat uncertain.

Conclusions

Neither the increased radiation from reactor-grade plutonium nor its increased critical mass prevent this material from being used to produce nuclear weapons. The gamma radiation from the cores of nuclear weapons made from reactor-grade plutonium can be easily shielded using a one centimeter layer of natural uranium. The processing of reactor-grade plutonium will not pose serious problems for new nuclear proliferants since these countries are unlikely to handle large amounts of reactor-grade plutonium each year. They also have the option to handle the plutonium remotely using computer controlled equipment, process the plutonium soon after chemical separation or simply expose their workers to higher amounts of radiation than U.S. standards would allow. That U 233, which can emit high doses of penetrating radiation, has been used to produce nuclear test devices is a clear indication that the increase radiation from reactor-grade plutonium will not pose a serious problem for its use in nuclear weapons. The critical mass of reactor-grade plutonium from high burnup LWR fuel is less than half that of HEU and therefore can be readily be used to produce nuclear weapons.

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[21] E. D. Clayton, "Anomalies of Nuclear Criticality, Revision 6," PNNL-19176, Pacific Northwest Laboratory, Richland, Washington, February 2010, pp. 108-112.

[22] Bare nuclear material not surrounded by any neutron reflecting substance. Neutron reflectors can substantially reduce the critical mass of some nuclear materials.

[23] E. D. Clayton, "Anomalies of Nuclear Criticality, Revision 6," PNNL-19176, Pacific Northwest Laboratory, Richland, Washington, February 2010, pp. 108-112; "Evaluation of nuclear criticality safety data and limits for actinides in transport," C4/TMR2001/200-1, Institut de Radioprotection et de Surete Nucleaire; Hemanth Dias, Nigel Tancock and Angela Clayton, "Critical Mass Calculations for ^{241}Am, ^{242m}Am and ^{243}Am," JAERI-Conf 2003-019; Rene G. Sanchez, *et.al.*, "Criticality of a ^{237}Np Sphere," Los Alamos National Laboratory, 2003; R. W. Brewer, "242 Pu Critical Mass," LA-UR-99-3509, Los Alamos National Laboratory.

Table 1

Unreflected Fast Critical Mass of Various Nuclear Materials

Nuclear Material	Critical Mass (Kilograms)	Directly Measured
Highly Enriched Uranium*	52.4	Yes
Weapon-Grade Plutonium** Delta phase	16.9	Yes
Reactor-Grade Plutonium*** Delta phase	19.3	Yes
Pu 238 Alpha phase	7 to 9	No
Pu 239 Alpha phase	10.0	Yes
Pu 240 Alpha phase	33 to 39	No
Pu 241 Alpha phase	12 to 13	No
Pu 242 Alpha phase	82 to 89	Yes
U 233****	16.2	Yes
Np 237	~ 60	Yes
Am 241	56 to 108	No

*93.7% U 235
**95.2% Pu 239, 4.5% Pu 240 and 0.3% Pu 241
***76.3% Pu 239, 20.2 % Pu 240, 3.1% Pu 241 and 0.4% Pu 242
****98.1% U 233, 1.3% U 234 and 0.6% U 238