

CHAPTER 6

Radiation and Critical Mass: No Barriers to Reactor-Grade Plutonium Use in Nuclear Weapons

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. This chapter demonstrates that neither of these factors poses a serious problem for the production of nuclear weapons using reactor-grade plutonium.

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 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 gamma

rays or x-rays. These radiations are far more penetrating than are alpha particles and can be a hazard to personnel though all of the x-rays and most 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.¹⁰⁸ At the same time the Pu-241 is also decaying into Am-241 and due to Am-241's long half-life, its quantity increases steadily for many years. Initially the gamma radiation contribution from U-237 is dominant, but in time

108. 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.

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 14.¹⁰⁹

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 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

109. 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.

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 half a centimeter of natural uranium. This would amount to less than 4 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 at least 100. Many of the weaker gamma rays from plutonium and Am-241 would be stopped completely. Therefore, by covering a sphere of high burnup plutonium with just one half a centimeter of uranium, the gamma dose would significantly less than the dose from an unshielded sphere of weapon-grade plutonium. Increasing the uranium covering to one centimeter (about 8 kilograms total) would increase the shielding by at least an additional factor of one hundred.

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 United States 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 5.8 kilogram sphere of reactor-grade plutonium,¹¹⁰ 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

110. 432 spontaneous fission neutrons produced per gram-seconds.

little to reduce this dose rate. However, at 1 meter away from the sphere, the dose rate would only be 2.4 mrad/hr. Even at this close distance, it would take over 2000 hours (about an entire year's worth of standard work weeks) to accumulate the 5 rem that is the U.S. standard for annual worker exposure to radiation.¹¹¹ There would be no need for military personnel to be that close to the finished plutonium weapons cores for such a long time.

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 five 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 United States developed its nuclear weapon production capacity in the 1940s and 1950s, there was little choice and most operations had to be performed 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.

111. For our purposes, the radiation units rad and rem are equivalent.

Another far cruder alternative would be for the worker exposure levels to be higher than what the United States 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.¹¹²

Some have asked that if reactor-grade plutonium can be used to produce nuclear weapons, why has the United States 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 United States 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.¹¹³ For the United States to change over to the use of reactor-grade

112. 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 available from <http://www.icrp.org/docs/2016tokyo/5%20Azizova.pdf>.

113. 10 CFR 20.1003.

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 weapon states 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 United States has had a nuclear test moratorium since 1992 which would prevent any such recertification. Since the United States has surplus weapon-grade plutonium, this is not an issue. However, for countries without access to weapon-grade plutonium but with access to large quantities of separated reactor-grade plutonium, the latter 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 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 ppm¹¹⁴ but techniques were developed that resulted in U-233 containing only about 5 to 10 ppm U-232.¹¹⁵

114. 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.

115. J. M. Boswell et. al., "Production of ²³³U with Low ²³²U Content," *Thorium*

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 the Th-228 decay products (thallium 208) emits a powerful gamma ray that is difficult to shield against.¹¹⁶ 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.¹¹⁷ U-233 containing 100 ppm U-232 emits 150 times as much gamma radiation as does reactor-grade plutonium. Even a one 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

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.

116. 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.

117. Jungmin Kang and Frank N. von Hippel, “U-232 and the Proliferation Resistance of U-233 in Spent Fuel,” *Science and Global Security* 9, 2001, Table 2, p. 10, available from <http://scienceandglobalsecurity.org/archive/sgs09kang.pdf>.

a weapons-usable material.”¹¹⁸ Presumably the U-233 would be managed by keeping personnel away from the cores most of the time. The United States and the Soviet Union are each known to have conducted at least one nuclear test using U-233.¹¹⁹

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 main isotope produced by this process is Np-237, which 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.

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% or less Pu-241. As a result the gamma radiation would be at least three times stronger from neptunium than from U-237 component of 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

118. Dean R. Tousley, Charles W. Forsberg, and Alan M. Krichinsky, “Disposition of Uranium-233,” ORNL/CP-95149, October 16, 1997.

119. David Holloway, “Research Note: Soviet Thermonuclear Development,” *International Security* 4, no. 3, Winter 1979/80, p. 195.

dose of the sphere to 300 mR/hr.¹²⁰ I calculate that even this small amount of shielding was able to reduce the gamma dose by about a factor of eight. Coating the neptunium with a half a centimeter of uranium would reduce the exposure from its gamma rays by a factor of about 200.

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.¹²¹ 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.¹²² Therefore, it is useful to review this issue.

120. Rene G. Sanchez, et.al., “Criticality of a ²³⁷Np Sphere,” Los Alamos National Laboratory, 2003.

121. Robert W. Selden, “Reactor Plutonium and Nuclear Explosives,” Lawrence Livermore National Laboratory, November 1976.

122. Bruno Pellaud, “Proliferation aspects of plutonium recycling,” *Journal of the Institute of Nuclear Material Management*, Fall 2002, p. 4.

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 neutron poisons. 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.¹²³ 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, 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.¹²⁴

The unreflected¹²⁵ critical masses of various nuclear materials relevant to the production of nuclear weapons are shown in Table 11.¹²⁶ Not all critical masses have been determined by direct mea-

123. S. R. Bierman and E. D. Clayton, "Criticality of Transuranium Actinides-Undermoderated Systems," *American Nuclear Society Transactions* 12, 1969. At that time data was only available for Pu-238 and Cm-244.

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

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

126. 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

surement 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 50% which is about half that of HEU. Such material could easily be used to produce nuclear weapons.

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 , $^{242\text{m}}\text{Am}$, and ^{243}Am ," JAERI-Conf 2003-019; Rene G. Sanchez, et.al., "Criticality of a ^{237}Np Sphere," Los Alamos National Laboratory, 2003; and R. W. Brewer, "242 Pu Critical Mass," LA-UR-99-3509, Los Alamos National Laboratory.

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

TABLE 11: Unreflected Fast Critical Mass of Various Nuclear Materials

*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

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.

In sum, 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 half centimeter layer of natural uranium. The neutron radiation is low enough so as to not have a serious effect on military personnel.

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 increased 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 about half that of HEU and therefore can be readily be used to produce nuclear weapons.