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

Part Four: Heat

This paper is the fourth 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

This paper will examine the issue of the heat produced by the decay of plutonium and how this heat might interfere with the production of a nuclear weapon. I will show that reactor-grade plutonium produced by high fuel burnup in current light water reactors (LWRs), by MOX fuel or recycled uranium can be effectively used in fission weapons using the U.S. early 1950s level of technology including a levitated design and modern high explosives.

Claims that the heat of plutonium from LWRs denatures that plutonium is based on faulty analysis that looks at only unlevitated nuclear weapon designs using World War II type explosives instead of levitated designs using more modern explosives. These claims also ignore techniques that allow the plutonium heat to be safely dissipated. These techniques include reducing the mass of plutonium in the weapon, using thermal bridges to conduct the heat away from the plutonium and using in-flight insertion of the plutonium so that it is only contained within the insulating high explosive shell for a short period of time. Further almost 50% of the large stocks of separated plutonium that exist worldwide are not nearly as hot as high burnup LWR fuel, having been produced in natural uranium fueled reactors or in reactors that use an enrichment lower than that used in LWRs.

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 <u>GregJones@proliferationmatters.com</u>

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. <u>Link</u>

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

Plutonium Decay Heat

All plutonium produces a significant amount of heat due to its radioactive decay. The plutonium that was used in the 1945 Trinity test consisted mostly of Pu 239 and was noticeably warm to the touch. The isotope Pu 238 is responsible for plutonium with a high heat output.⁵ It produces over 200 times as much heat as does Pu 239. Pu 238's heat dominates the heat output of any plutonium that is more than about 0.5% Pu 238, though for plutonium to have a high heat output it must contain at least several percent Pu 238. Pu 239 as well as the higher plutonium isotopes Pu 240, Pu 241 and Pu 242 have their origin in an initial neutron capture in U 238. Pu 238, however, has its origin in an initial neutron capture in U 235 and requires additional neutron captures in U 236 and Np 237. As a result not much Pu 238 is produced in natural uranium fuel where the initial U 235 content is low as is the burnup. High Pu 238 plutonium is produced in enriched uranium fuels with a high initial U 235 content and high burnups. It can also be produce in MOX fuel (fuel that initially contains both plutonium and uranium) or recycled uranium fuel that already contains some U 236. There have been proposals to produce plutonium with a very high Pu 238 content by spiking enriched uranium fuel with either neptunium or americium 241. This has never been done, nor is it likely to be done, since this would increase the fuel cost and make the fuel more difficult to handle. Table 1 (Appendix) shows the heat output of plutonium produced by different types of reactors with different burnups.

For decades it was suggested that the high heat from Pu 238 would denature plutonium in cases where the Pu 238 content is several percent. Until recently, there was never any specific analysis to delineate how high the Pu 238 content would have to be to result in denatured plutonium. However, Gunther Kessler, a proponent of the false notion of denatured plutonium, has produced some specific analysis.⁶ Kessler's analysis suggests that plutonium with a heat output of about 15 watts per kilogram would melt the high explosives that were used in World War II and plutonium with a heat output of about 110 watts per kilogram would be enough to melt the center of the plutonium core.

Plutonium Weapon Core

The plutonium itself is not going to be a constraint on the acceptable amount of heat from plutonium. Though some still sometimes believe that the plutonium in nuclear weapons is alpha phase which is quite sensitive to heat, it is now well known that plutonium in nuclear weapons is a plutonium alloy containing one percent by weight (3.2 atom percent) of gallium which stabilizes the plutonium in the delta phase.⁷ The alloy is very heat resistant and is stable from room temperature to over 500 degrees centigrade. It has a very low coefficient of expansion over this range.⁸ Above about 530 degrees centigrade the plutonium transitions from delta phase to

⁵ For the decay heat of different plutonium isotopes see: Gregory S. Jones, "The Myth of "Denatured" Plutonium, Reactor-Grade Plutonium and Nuclear Weapons, Part One, July 26, 2016, Table 1. <u>Link</u>

⁶ G. Kessler, *Proliferation-Proof Uranium/Plutonium Fuel Cycles: Safeguards and Non-Proliferation*, KIT Scientific Publishing, 2011.

⁷ For a discussion of plutonium phases see: Gregory S. Jones, "Fissile Material Conversion Times, Wastage and Significant Quantities: Lessons from the Manhattan Project," December 16, 2015 <u>Link</u>

⁸ Siegfried S. Hecker, "Plutonium and Its Alloys," *Los Alamos Science*, No. 26, 2000, p 293.

epsilon phase and contracts significantly which could damage the plutonium core in a nuclear weapon.

By Kessler's own calculations, achieving a temperature of over 530 degrees centigrade would require plutonium that had a heat output of about 60 watts per kilogram. By simply lowering the amount of plutonium in the device, this limit could be raised to 100 watts per kilogram. Kessler assumes that the plutonium in a nuclear weapon is near critical and that the weapon contains about 10 kilograms of reactor-grade plutonium so that the total plutonium heat output would be about 600 watts. As I showed in my last paper, quite satisfactory nuclear weapons can be produced using just 6 kilograms of reactor-grade plutonium.⁹ If 6 kilograms of plutonium is made into a shell having the same outer diameter as 10 kilograms of plutonium, then the 6 kilograms of plutonium could have a heat output of 100 watts per kilogram and match the outer temperature of a 10 kilogram plutonium sphere with a heat output of 60 watts per kilogram. Reactor-grade plutonium with such a high heat output does not exist and likely never will.

Plutonium From Nuclear Reactors Using Natural or Slightly Enriched Uranium Fuel

Even if Kessler were correct regarding the proper limits for the heat from plutonium, as can be seen from Table 1 (Appendix), significant amounts of plutonium are far cooler than the Kessler's 15 watts per kilogram limit for World War II type explosives. Most notable is the plutonium produced by power reactors fueled with natural uranium (CANDU and MAGNOX). Currently there are 46 power reactors in operation fueled with natural uranium.¹⁰ The majority of these are in either Canada or India but there are some in Argentina, China, Pakistan, Romania and South Korea. At the present time, only the spent fuel from the reactors in India is being reprocessed but the spent fuel from these other reactors could also be reprocessed.

All of the current natural uranium fueled power reactors are heavy water reactors (CANDU) but in the past there were 38 natural uranium fueled graphite power reactors (MAGNOX). Twenty six of these reactors were in the UK, nine were in France and one each in Spain, Italy, and Japan. The last of these reactors operated in the UK and was shut down in December 2015. All of the spent fuel from these reactors has been or is going to be reprocessed. Due to its low heat and Pu 241 content (a source of radiation exposure), this plutonium is preferred for the production of MOX fuel. As a result, it is likely that most if not all of the plutonium produced by the reactors in France, Spain, Italy and Japan has been consumed as MOX fuel. This is not the case for the plutonium produced in the UK, where most if not all of the plutonium produced by the natural uranium fueled graphite reactors (roughly 70 metric tons) is being stored as part of the UK's massive plutonium stockpile.

In addition, there are nuclear power reactors which use low enriched uranium fuel where the initial enrichment is significantly less than that used in modern LWRs and the burnup is less as well. One group of such reactors is the 14 advanced gas cooled reactors (AGRs) in operation in

⁹ Gregory S. Jones, "The Myth of "Denatured" Plutonium, Reactor-Grade Plutonium and Nuclear Weapons, Part Three: Predetonation," October 25, 2016. <u>Link</u>

¹⁰ There is also a heavy water nuclear power reactor in Argentina (Atucha 1) which uses 0.9% enriched uranium fuel. The plutonium produced by this reactor is only slight hotter than that produced in a natural uranium fueled reactor.

the UK. The initial enrichment of the fuel for these reactors is only about 2.5% and the burnup is about 18,000 MWD/Te. Up to now the spent fuel from these reactors has all been reprocessed (though this may end in the next few years) and has resulted in about 40 metric tons of plutonium whose the heat output is about 7 watts per kilogram (see Table 1, Appendix). Therefore at least 110 metric tons of the massive stockpile of separated plutonium stored in the UK¹¹ is low heat plutonium. The UK has no plans for the disposal of this plutonium.

Another group of reactors similar in fuel enrichment and burnup to the AGRs are the RBMK reactors, all of which were built in the Soviet Union. The Chernobyl reactor was an RBMK. Today 11 such reactors are operating, down from a total of 17. Of Russia's current civil stockpile of separated plutonium of 52 metric tons, roughly 20 metric tons were produced in this type of reactor. Combined with the approximately 110 metric tons of separated plutonium in the UK, this means that roughly 130 metric tons of the 271 metric ton world stockpile of separated civil plutonium (almost 50%) has a heat output well below Kessler's 15 watt per kilogram limit.

Even if one believed that high heat plutonium was denatured, what should be done about the 72 nuclear power reactors that do not produce high heat plutonium? Should they all be shut down? Proponents of the notion that heat can denature plutonium are silent on this issue.

Nor do all LWRs necessarily use high initial fuel enrichment and high fuel burnup (for example 4.3% enriched with a burnup of 51,000 MWD/Te) resulting in plutonium with a high heat output. Iran's Bushehr LWR, which started operation in 2012, uses fuel with an initial enrichment of only 3.6% with a full burnup of 37,000 MWD/Te.¹² The plutonium produced in such fuel would have a heat output of about 13 watts per kilogram which is still less than Kessler's 15 watts per kilogram limit. The Bushehr reactor is one of the LWRs with the highest concerns regarding proliferation but the use of lower initial fuel enrichment and lower full burnup could apply to any LWR since there is no requirement that such reactors use the highest initial fuel enrichment possible.

Even for LWRs that use uranium fuel with a high initial enrichment, not all of the fuel will have a high burnup. Most notably, when any LWR starts initial operation, it will use some fuel whose enrichment is well below its normal enrichment. When a reactor has been in sustained operation, it contains a mixture of fuel with different burnups. But when a reactor starts for the first time, it must use fuel with different levels of enrichment. Fuel with the lowest enrichment is burned for only a relatively short time before being permanently discharged. This first discharge fuel will contain plutonium which may not even be reactor-grade but rather fuel-grade. Its heat output will be no more than 3 to 4 watts per kilogram.¹³ This is similar to plutonium produced by full burnup natural uranium fuel. It is certainly not denatured by heat, since its heat output is far less than Kessler's 15 watt per kilogram limit. This first reactor discharge might contain close to 100 kilograms of plutonium, enough for at least 15 nuclear weapons.

¹¹ At the end of 2015 the total stockpile of separated plutonium in the UK was 129 metric tons. See "Annual figures for holdings of civil unirradiated plutonium as at 31 December 2015," UK Office for Nuclear Regulation.

¹² Gregory S. Jones, "Iran's Bushehr Nuclear Power Reactor: A Potential Source of Plutonium for Nuclear Weapons," March 24, 2016. <u>Link</u>

¹³ *Ibid*.

Low burnup is not necessarily an issue only when a power reactor starts operation for the first time. Even when operating normally, some reactor fuel from modern LWRs is discharged with less than full burnup. It would be easy for a country to claim some technical fault in reactor fuel and discharge the fuel with far less than full burnup. This plutonium could be fuel-grade or even weapon-grade. Iran temporarily discharged the entire fuel core from the Bushehr reactor during reactor testing in 2012. The reason for this discharge was never explained. The International Atomic Energy Agency might detect such early discharge but would have no reason to declare a safeguards violation.

High Heat Plutonium is Not Denatured

Even when plutonium is produced by high burnup enriched uranium fuel in LWRs, the heat will not denature the plutonium. As was discussed above, the easy expedient of using a reduced plutonium mass in the weapon would allow high heat plutonium to be used in a simple nuclear weapon without melting the high explosive. If 6 kilograms of plutonium were used in a weapon instead of Kessler's 10 kilograms, then Kessler's 15 watt per kilogram limit would become 25 watts per kilogram (15 x 10/6). As can be seen from Table 1(Appendix), this is significantly higher than the 17 watt per kilogram heat output of plutonium produced by high burnup enriched uranium LWR fuel. Since almost all of the world's current stockpile of separated plutonium produced in LWRs was produced using enriched uranium fuel uncontaminated by U-236 (i.e. did not use recycled uranium), this means that all this plutonium is not denatured by its decay heat.

But what about plutonium produced in MOX fuel or in enriched uranium fuel that has been contaminated by high levels of U-236 (recycled uranium)? Plutonium produced from these fuels might have a heat output in the range of 30 to 40 watts per kilogram. The first point to note is that up to now very little plutonium has been separated from these types of fuels. This is unlikely to change in the future.¹⁴ The high heat (and the relatively high radiation) from this plutonium make it undesirable for use as MOX fuel and indeed the characteristics of this plutonium may exceed what current MOX fabrication plants are licensed to handle.

Second, Kessler's 15 watt per kilogram limit applies only to World War II type explosives and more modern explosives are less sensitive to heat. World War II explosives might melt at temperatures of less than 100 degrees centigrade, whereas more modern explosives might not melt until 190 degrees centigrade. In addition, more modern explosives have somewhat better heat transfer characteristics.

Consider the case of 6 kilograms of reactor-grade plutonium with a heat output of 40 watts per kilogram. The total heat output is 240 watts. Kessler has performed a calculation for a case where the core has a total heat output of 240 watts for a weapon using modern high explosives (his "medium technology" case). He finds that the inner edge of the high explosive layer would have temperature of about 240 degrees centigrade, higher than 190 degrees centigrade. Kessler then concludes that such a nuclear weapon could not function.

¹⁴ France, the only country to recycle uranium in a significant way, stopped in 2010, in part because the French utility (EDF) objected to the high cost. See: International Panel on Fissile Materials, "Plutonium Separation in Nuclear Power Programs," 2015, p. 34.

But Kessler's calculation is based upon a solid pack nuclear weapon design where every layer of the weapon is in contact with the next layer. However, U.S. nuclear weapons of the early 1950s used "levitation" where a void is introduced into weapon (i.e. there is an empty space is between two of the layers) to improve weapon performance. From their weight and yield, it appears that even 50 years ago, the first French and Chinese nuclear weapons employed levitation.

If one uses a 10 centimeter void in Kessler's design, expanding the outer shell of the weapon by this amount, then there is a dramatic temperature drop. The inner edge temperature of the high explosives layer would only be about 140 degrees centigrade, well below 190 degrees centigrade, and there should be no problem with the functioning of the weapon.¹⁵ Therefore, simply by reducing the mass of plutonium, using a levitated design and modern high explosives, it is quite possible to use reactor-grade plutonium with a heat output of at least 40 watts per kilogram. This heat output exceeds that of plutonium produced in MOX fuel or plutonium produced by recycling uranium.

Nor are these the only techniques to deal with high heat plutonium. J. Carson Mark has suggested using an aluminum thermal bridge to conduct heat away from the plutonium core which could result in halving the plutonium core temperature.¹⁶ Simple calculations show that the aluminum segments running through the high explosives would be less than one tenth of one millimeter thick, which would be unlikely to interfere with the functioning of the high explosives. Therefore the use of a thermal bridge might allow the acceptable heat level of plutonium for nuclear weapons to be as high as 80 watts per kilogram. Various proponents of the concept of denatured plutonium have suggested that the aluminum running through the high explosive implosion system would interfere with the weapons functioning. They have made these claims even though they have no background in nuclear weapon design, unlike Mark who was Director, Theoretical Division, Los Alamos National Laboratory from 1947 to 1972.

Further, in the early 1950s the U.S. did not store its plutonium cores inside the high explosives, but rather stored them separately for safety and security reasons. Pakistan is reported to use the same system today. U.S. 1950s era weapons used in-flight insertion where the plutonium core was only inserted into the high explosive assembly after the weapon was in flight, meaning that it occurred only minutes before detonation. Using this method there would be no long-term exposure of the high explosives to the heat (or radiation) of the plutonium core.

Therefore, there are a number of ways that high heat plutonium could be used in simple unboosted implosion designs of the type that proliferants might develop early in their nuclear weapon program. This observation is confirmed by U.S. statements that Am-241 could be used to produce nuclear weapons.¹⁷ Its heat output is 114 watts per kilogram, significantly higher than that of any plutonium.

¹⁵ The lower temperature is achieved because with a larger diameter, the high explosive shell has a larger surface area. For the case where the plutonium has a heat output of 20 watts per kilogram, the inner edge temperature of the high explosives layer would be less than 90 degrees centigrade. As noted, almost all separated plutonium, including that produced by high burnup in LWRs has a heat output of less than 20 watts per kilogram.

 ¹⁶ J. Carson Mark, "Explosive Properties of Reactor-Grade Plutonium," *Science & Global Security*, Vol. 4, 1993.
¹⁷ David Albright and Kimberly Kramer, "Neptunium 237 and Americium: World Inventories and Proliferation

Concerns, June 10, 2005, revised August 22, 2005.

Conclusions

Plutonium decay heat even from plutonium with a high Pu 238 content is not an impediment to the use of this plutonium in simple unboosted implosion nuclear weapons. By using a reduced plutonium core mass in a levitated weapon design utilizing modern high explosives would allow the use of plutonium with a heat output of 40 watts per kilogram, a higher heat output than that produced even in MOX fuel or fuel using recycled uranium. The use of conductive aluminum bridges through the high explosive is another technique that could raise the acceptable level of plutonium decay heat to as high as 80 watts per kilogram.

Almost all separated plutonium is much cooler than even 40 watts per kilogram. Plutonium from uranium fueled LWRs that do not use recycled uranium (almost all of the LWRs in the world) has a heat output of less than 20 watts per kilogram and in some cases less than 10 watts per kilogram. The plutonium from AGRs and RMBK type reactors has a heat output of about 7 watts per kilogram. The plutonium from natural uranium fueled reactors has a heat output of less than 4 watts per kilogram. Since the plutonium cores can be kept separate from the high explosive assembly until minutes before the weapon is detonated, the exposure of the high explosives to the heat and radiation of the plutonium core can be minimized. It is time to lay to rest the notion that heat can denature plutonium.

Appendix

Table 1

Spontaneous Fission Neutrons and Decay Heat of Plutonium Produced in Different Types of Reactors with Different Burnups¹⁸ (Ten Years After Discharge)

Plutonium	Pu-238%	Pu-239%	Pu-240%	Pu-241%	Pu-242%	Spontaneous	Decay
Reactor						Fission	Heat
Type and						Neutrons	(watts per
Burnup						(neutrons	kilogram)
(MWD/TE)						per gram-	
						seconds)	
Weapon-		93.4	6.0	0.6		55	2.2
Grade							
CANDU	0.07	69.0	26.5	3.1	1.3	265	3.6
7,060							
MAGNOX	< 0.1	69.9	25.5	3.4	1.2	254	3.6
5,000							
AGR	0.6	55.8	32.0	6.3	5.2	395	6.9
18,000							
LWR	0.1	77.8	18.1	3.5	0.5	176	3.4
1^{st}							
Discharge							
LWR	0.6	69.8	20.6	6.9	2.2	240	6.4
20,000							
LWR	1.3	58.8	25.9	8.7	5.4	361	10.5
33,000							
LWR	2.6	54.3	25.8	9.7	7.6	432	17.8
51,000							
LWR	3.3	41.3	33.0	10.7	11.6	583	22.0
MOX							
51,000							
LWR	6.3	61.5	19.4	8.8	4.0	408	38.1
Recycled U							
46.300							

¹⁸ For more information on the entries in this table see: Gregory S. Jones, "The Myth of "Denatured" Plutonium, Reactor-Grade Plutonium and Nuclear Weapons, Part One" July 26, 2016. <u>Link</u>