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The History of the Pu 240 Content of U.S. Weapon-Grade Plutonium

Introduction and Summary

Ideally plutonium used in nuclear weapons would contain only the isotope Pu 239. However, the U.S. discovered in 1944 that plutonium produced in nuclear reactors must contain at least a small percentage of the isotope Pu 240. The relatively high spontaneous fission rate of Pu 240 causes this isotope to release neutrons which can cause unboosted nuclear weapons to have a significant probability of predetonating i.e. starting the nuclear reaction prematurely, resulting in a lower than designed nuclear yield. Therefore limits were placed on the percentage of Pu 240 in the plutonium used in early U.S. nuclear weapons so as to ensure a reasonable probability of achieving the design yield. Plutonium that the U.S. uses in its nuclear weapons is termed "weapon-grade."

Currently, the U.S. defines weapon-grade plutonium as having a Pu 240 content of less than 7%[2] and U.S. nuclear weapons use plutonium with a Pu 240 content of about 6%. Given that all U.S. nuclear weapons are now boosted, this limit on Pu 240 content has nothing to do with the probability of predetonation but rather with other properties of the plutonium such as radiation output. But this was not always the case. The U.S. did not deploy boosted fission weapons until 1957 and continued to stockpile unboosted nuclear weapons well into the 1960s.[3]

The declassification of documents related to the operation of the plutonium production reactors at Hanford allows the construction of a history of the Pu 240 content of U.S. weapon-grade plutonium. In the 1940s the limit on the permissible Pu 240 content was fairly low due to the relatively slow assembly time associated with early implosion fission weapons. As implosion technology improved, the Pu 240 limit increased.

The Nagasaki weapon used plutonium that was only 1.0% Pu 240.[4] Soon after the Nagasaki weapon was employed, the limit was increased to 2.0% Pu 240. In 1949 the limit was increased to 3.8% and in March 1951 to 5.5%. This high limit is an indication that even in 1951, implosion fission technology had been significantly improved over that used in the Nagasaki weapon. Indeed in the 1950s, U.S. implosion fission weapons employed a technique known as levitation, which is the use of an air gap between the weapon's fissile core and the tamper. This air space allows the implosion wave to increase in speed and compress the nuclear core more rapidly.

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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, this paper is not related to any RAND project and therefore RAND should not be mentioned in relation to this paper. I can be reached at GregJones@proliferationmatters.com

[2] *Plutonium: The First 50 Years*, DOE/DP-0137, U.S. Department of Energy, February 1996, p.17.

[3] N. Stetson et al., *Savannah River Production Reactor History*, CIV-685-2A, September 1963.

[4] Gregory S. Jones, "Fissile Material Conversion Times, Wastage and Significant Quantities: Lessons from the Manhattan Project," December 16, 2015 p.10.

In 1954 the Pu 240 limit for most weapon-grade plutonium being produced was 8.8%. It is interesting that the limit was so high given that this plutonium would have been used in the unboosted implosion fission weapons used as the fission triggers (primaries) in early U.S. two-stage thermonuclear weapons. In such weapons, a dependable high primary yield is necessary to ensure the proper functioning of the thermonuclear stage.

However, Hanford never produced weapon-grade plutonium with a Pu 240 percentage this high since operating problems at Hanford not the neutron output of the plutonium, determined the Pu 240 content of the plutonium. Due to the fuel rupture problem, Hanford was only able to produce plutonium that was 5.5% Pu 240 and in 1957 and 1958 actually had to reduce the fuel burnup, which resulted in a Pu 240 content of 4.7%.

With the advent of tubular fuel elements in 1959, the limit was increased to 6.0% and it soon became frozen at this level. It is interesting to speculate what might have been the result if Hanford had been able to produce plutonium that was 8.8% Pu 240 in the mid-1950s. Perhaps this Pu 240 percentage would have become the standard and all U.S. weapon-grade plutonium today would have a Pu 240 content of 8.8%.[5] Table 1 gives a breakdown of my estimates of the amounts of weapon-grade plutonium produced at Hanford with various Pu 240 contents and the dates and reactors involved in its production.

Table 1
Amounts of Weapon-Grade Plutonium Produced at Hanford:
Pu 240 Content, Dates Produced & Reactors Used

Pu 240 Content*	Amount in Metric Tons	Dates Produced and Reactors Used
2.0%	1.7	1945-1948, B, D & F 1954-1956, C
3.8%	0.7	1949-1951 B, D, DR, F & H
4.7%	6.0	1957-1958 All reactors except N
5.5%	5.3	1951-1956 All reactors except N and C in 1954-1956
6.0%	40.8	1959-1971 & 1983-1987 All reactors
5.7% Weighted Average	54.5 Total	

*Before 1961 the Pu 240 content varied significantly from batch to batch.

[5] Note that the U.S.-Russian 2000 Plutonium Management and Disposition Agreement defines weapon-grade plutonium as having no more than about 9.1% Pu 240 (a Pu 240 to Pu 239 ratio of no more than 0.1).

History of the Production of Weapon-Grade Plutonium at Hanford

The declassification of many documents regarding the plutonium production operations at Hanford provides much information but using these documents also presents some difficulties. Only some of the documents produced by Hanford have been declassified, so it is sometimes difficult to place a document in the proper context. Also the documents use a good deal of jargon (such as "E-metal," i.e. uranium fuel enriched to 0.95%) whose meaning was obvious at the time but not so today.

The meaning of some technical terms is not always clear. An important term for this paper is MWD/ton (Megawatt Day per ton), which is the measure of fuel burnup. For a number of decades it has been standard to measure burnup in terms of MWD/te, where the "te" is a metric ton (2,205 lb). In most of the Hanford documents it is not obvious what kind of ton is meant. It would be tempting to assume that they meant metric tons but, in fact, they are short tons (2,000 lb.).

Finally, due to the limitations of the time, some of the information was simply incorrect. Of great importance for this paper is Hanford's estimate of the Pu 240 content of the plutonium it was producing. But in the 1950s Hanford was not measuring this directly but rather measuring the property that was actually important for weapons use, namely the plutonium's neutron production in units of n/g-s (neutron per gram seconds). Hanford then converted this neutron measurement into a Pu 240 content by using the neutron production rate of Pu 240. But Hanford's estimate of the Pu 240 neutron production rate in the 1950s was too high by about 30 percent, which means that its estimate of the Pu 240 content of any given plutonium was about 30 percent too low.[6]

Table 2 shows the operating history of the Hanford plutonium production reactors. The B, D and F reactors were built during World War II. Soon after the war it was discovered that the operation of the reactors was causing their graphite moderator to expand to such an extent that it threatened the continued operation of the reactors. The B reactor was shut down to preserve some of its operating life and the H reactor started construction. The situation at the D reactor was so serious that the DR reactor was built to replace it. However, a solution was found to the graphite problem and the D reactor was never shut down.[7]

These first 5 reactors (B, D, F, H, & DR) had an identical design. The C reactor was a slightly improved design. The KW and KE reactors were improved designs with a significantly higher power level and conversion ratio. The N reactor used enriched uranium fuel and was designed to produce electricity as well as plutonium. Most of the plutonium produced by this reactor was not weapon-grade due to the higher burnup of its enriched uranium fuel.

[6] Compare Figure 13 (p.42) from F. E. Kruesi, J. O. Erkman, and D. D. Lanning , "Critical Mass Studies of Plutonium Solutions," General Electric, Richland Washington, May 19, 1952, HW-24514 with the more accurate Figure 6 (p.32) "Hanford Reactor and Separations Facility Advantages," Hanford Atomic Products Operation, Richland Washington, June 27, 1963, HW-78100.

[7] It was found that heating the graphite annealed the damage caused by irradiation.

To a first approximation the amount of plutonium produced is directly proportional to the power level of a reactor. As can be seen from Table 2 the power level of the earliest reactors was increased by nearly a factor of ten over their operating life. This was achieved in steps over time by allowing higher water discharge temperatures, increasing reactor cooling capacity and providing small amounts of enriched uranium. The reactors at Hanford produced a total of about 54.5 metric tons of weapon-grade plutonium.[8] The annual production peaked at over 4 metric tons between 1960 and 1965 and was essentially over by 1971.[9]

As was discussed above, the plutonium in the Nagasaki weapon had a Pu 240 content of 1.0%. But even before the Nagasaki weapon had been used in combat, General Groves, the head of the Manhattan Project, reported that the Pu 240 content of the plutonium was going to be increased.[10] In August "the customer" requested that the reactor fuel burnup be limited to 200 MWD/ton which would result in a Pu 240 content of about 2.0%.[11] An operating report from November 1945 refers to 200 MWD/ton as "normal discharge material."[12] Apparently this fuel burnup goal lasted until about the end of 1948. Between 1944 and the end of 1948 Hanford produced 676 kilograms of plutonium.[13]

Table 2
Operating History of the Hanford Plutonium Production Reactors

Reactor	Operating Dates	Design Power Level MWt	Highest Sustained Power Level MWt
B	9/44-3/46 7/48-2/68	250	2210
D	12/44-6/67	250	2165
F	2/45-6/65	250	2040
H	10/49-4/65	400	2140
DR	10/50-12/64	250	2015
C	11/52-4/69	650	2500
KW	1/55-2/70	1850	4400
KE	4/55-1/71	1850	4400
N	12/63-1/87	4000	4000

At the beginning of 1949 the fuel burnup discharge goal was raised to 400 MWD/ton which is a Pu 240 content of about 3.8%. During the first half of 1949 the fuel burnup was gradually raised

[8] *Plutonium: The First 50 Years*, DOE/DP-0137, U.S. Department of Energy, February 1996, pp. 28-29.
[9] The N reactor produced 2,778 kilograms of weapon-grade plutonium between 1983 and 1987. *Ibid.*
[10] General Leslie Groves, "Memorandum to the Chief of Staff," July 30, 1945, Manhattan Engineering District Papers, Box 3, Folder 5B, Record Group 77, Modern Military Records, National Archives, Washington, D.C.
[11] W.O. Simon, "Hanford Engineer Works monthly report, August 1945," September 9, 1945 HW-7-2361-Del.
[12] W.E. Jordan "100 Area Technical Activities Report-Physics, 10/25/45 to 11/25/45," November 29, 1945, E.I. Du Pont De Nemours & Company.
[13] *Plutonium: The First 50 Years*, DOE/DP-0137, U.S. Department of Energy, February 1996, pp. 28-29.

from 200 MWD/ton to 400 MWD/ton.[14] It continued at this level until March 1951. During this time Hanford produced 734 kilograms of plutonium.[15]

In March 1951 the burnup goal was raised to 600 MWD/ton which yields a Pu 240 content of about 5.5%.[16] Apparently the burnup was raised to this new level almost immediately. Unfortunately, this burnup increase lead to an immediate increase in the number of fuel ruptures.

Fuel ruptures were a major concern for the operations at Hanford. When a fuel element ruptured the hot metallic uranium was exposed to the water coolant. It would oxidize and expand blocking the fuel channel. This would cut off the flow of coolant to the other fuel elements and in the worst case, these fuel elements could catch fire and set the entire reactor ablaze. Therefore there were systems that quickly detected any fuel rupture. Once detected, the reactor would be shut down and the ruptured element removed.

In the best case, the ruptured element could be removed in just a half an hour. In the worst case, the fuel element would be stuck and so much force would be required to remove it that the aluminum fuel channel would be damaged and would have to be replaced. Or the swollen ruptured fuel element could rupture the fuel channel leading large amounts of water to spill into the reactor. The reactor's graphite would then need to be dried before the reactor could be restarted. In either case, days of reactor operation could be lost to a fuel rupture. In optimizing the plutonium production operations at Hanford, the likelihood of fuel ruptures needed to be taken into account. Since this likelihood increased with fuel burnup as well as reactor power level, the threat of fuel rupture tended to limit the fuel burnup and thereby the Pu 240 content of the plutonium produced.

By improving fuel quality, it was possible to reduce the fuel rupture rate and maintain the 600 MWD/ton goal at the five oldest reactors (the B, D, F, H and DR) through the end of 1956. However, when the new C reactor came on line at the end of 1952, its higher power level and thereby higher power density resulted in a large number of fuel ruptures. Since there was a new requirement from the U.S. Atomic Energy Commission (USAEC) for some low Pu 240 plutonium, the C reactor was operated with a fuel burnup goal of about 200 MWD/ton (a Pu 240 content of about 2.0%) from 1954 through the end of 1956. During this time I estimate that about 1.0 metric ton of plutonium was produced by the C reactor. Hanford produced a total of 6,320 kilograms of plutonium between March 1951 and the end of 1956.[17] Subtracting the approximately one metric ton of plutonium produced by the C reactor between 1954 and 1956 gives a total of about 5.3 metric tons of plutonium with a 5.5% Pu 240 content produced by the other reactors at Hanford between March 1951 and the end of 1956.

A 1954 document reveals the specific maximum plutonium neutron output in terms of n/g-s values that were required by the USAEC.[18] For the low burnup plutonium its n/g-s should not exceed 20 which would be a Pu 240 content of about 2.2%. For all other weapon-grade

[14] Various P Division monthly reports.
[15] *Plutonium: The First 50 Years*, DOE/DP-0137, U.S. Department of Energy, February 1996, pp. 28-29.
[16] DL Deneal, "Historical Events-Single Pass Reactors and Fuels Fabrication," April 10, 1970, DUN-6888, p.7.
[17] *Plutonium: The First 50 Years*, DOE/DP-0137, U.S. Department of Energy, February 1996, pp. 28-29.
[18] R. O. Gumprecht, "Plutonium Product Quality," May 28, 1954, HW-31952, General Electric, Richland Washington.

plutonium its n/g-s should not exceed 80 which would be a Pu 240 content of about 8.8% (a burnup of about 1,050 MWD/ton). The production of tritium-filled reservoirs did not begin until 1957, meaning that the U.S did not have boosted weapons until then. Therefore plutonium with a Pu 240 content of 8.8% was considered acceptable in unboosted implosion fission weapons. Unfortunately there is no indication of when these plutonium requirements came into effect or how long they lasted.

Apparently in 1954 and 1955 Hanford tried to increase the fuel burnup to levels higher than 600 MWD/ton but fuel ruptures stymied this effort. In the mid-1950s it was the fuel rupture rate, not the plutonium neutron output that determined the Pu 240 content of the plutonium produced.[19] As the power levels of the reactors continued to increase, the fuel ruptures increased as well. As a result, in 1957 and 1958 the fuel burnup was reduced to 500 MWD/ton (a Pu 240 content of about 4.7%). Hanford produced 5,965 kilograms of plutonium during these two years.[20]

In 1959 with the advent of tubular fuel elements (I & E fuel elements in Hanford jargon) that were less rupture prone, the fuel burnup was increased to about 675 MWD/ton (a Pu 240 content of about 6.0%). In early 1961 Hanford was given an explicit Pu 240 goal of 6.0%, instead of the goal being set in terms of fuel burnup. Further this new goal had to apply to all the plutonium produced whereas in the past there had been significant variation in the Pu 240 content from batch to batch as Hanford optimized the reactor operations to maximize plutonium output. This new goal caused Hanford some concern since it would was difficult to convert this requirement into a fuel burnup (it was difficult to keep track of the fuel burnup in each fuel channel).[21] In the end, it appears that Hanford had no trouble meeting this goal.

There is no indication that there were any further changes to the Pu 240 requirement for weapon-grade plutonium. By the mid-1960s the plutonium production declined as various reactors were shut down and some plutonium was produced for non-weapon purposes. Weapon-grade plutonium production at Hanford ended in 1971 with the shutdown of the KE reactor.

The N reactor continued in operation until 1987. However, most of the plutonium this reactor produced was for non-weapon purposes though it did produce 2,778 kilograms of weapon-grade plutonium between 1983 and 1987.[22] From 1959 through 1987, when the Pu 240 specification was 6.0%, Hanford produced 40,772 kilograms of plutonium. [23] This was about 75% of Hanford's total weapon-grade plutonium production.

Much less is known about the plutonium production at Savannah River since far less has been declassified about the reactor operations there. However, of the 36.1 metric tons of weapon-grade plutonium produced there, 32.5 metric tons (90 percent of the total) were produced after 1958. Therefore it is safe to say that the vast majority of the weapon-grade plutonium produced at Savannah River had a Pu 240 content of 6.0%.

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[19] R. A. Pugh, "Final Report-Production Test 200-2 Processing of Special Irradiated Plutonium," September 27, 1956, HW-45940, p.4.
[20] *Plutonium: The First 50 Years*, DOE/DP-0137, U.S. Department of Energy, February 1996, pp. 28-29.
[21] See: L. W. Lang & W. I. Neef, "Notes on Reactor Operation within a Product Specification," June 9, 1961, HW-69904 and T. Prudich, "Product Quality and an Interim Goal Exposure Plan," June 12, 1961, HW-69912.
[22] *Plutonium: The First 50 Years*, DOE/DP-0137, U.S. Department of Energy, February 1996, pp. 28-29.
[23] *Ibid.*

Table 1 in the summary gives a breakdown of the amounts of weapon-grade plutonium produced at Hanford with various Pu 240 contents. Until 1951 the Pu 240 content was determined by the specifications for plutonium neutron output. Between 1951 and 1959, it appears that fuel ruptures, not plutonium neutron output, determined the Pu 240 content of the plutonium.

Though at one time plutonium with a Pu 240 content of 8.8% was acceptable from a neutron output standpoint, Hanford could not efficiently produce such plutonium. If Hanford had been able to produce large amounts of plutonium that was 8.8% Pu 240, such plutonium might have become the standard and all U.S. weapon-grade plutonium today might have an 8.8% Pu 240 content. It is interesting to note that the U.S.-Russian 2000 Plutonium Management and Disposition Agreement defines weapon-grade plutonium as having a Pu 240 content of no more than about 9.1% (a Pu 240 to Pu 239 ratio of no more than 0.1).

Since the U.S. did not deploy boosted fission weapons until 1957, this means that 8.8% Pu 240 plutonium could have been used in the unboosted implosion fission weapons deployed in the 1950s. This could have included early U.S. two-stage thermonuclear weapons since such weapons used unboosted implosion fission weapons as their primaries. This indicates that in the early to mid-1950s, the U.S. had already achieved implosion fission weapons that were significantly more advanced than the Nagasaki weapon.