Plutonium from Light Water Reactors as Nuclear Weapon Material

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The feasibility of using "reactor grade" plutonium (Pu) for nuclear explosives was established some years ago. The Department of Energy, in 1977, declassified the fact that an underground test had been conducted (in 1962) in which weapon grade Pu had been replaced with reactor grade Pu with successful results. They re-emphasized the test in 1997 for the arms control community[1]. The subject of illegal construction of nuclear explosives was reviewed by J Carson Mark, late T-Division head at Los Alamos National Laboratory (LANL), in a 1990 report.[2] The focus of his report was terrorist organizations with access to spent reactor fuel, and he concluded that the difficulties encountered by a terrorist group in using reactor grade Pu for explosive fabrication differ only in degree, but not in kind, from the problems they would find using weapon grade Pu. In this note, however, we are more interested in a "rogue" state with access to Pu from a light water reactor (LWR), and having perhaps more technical expertise, and certainly more resources available, than the putative terrorist group. Our approach is qualitative with quantification of key items necessary for estimates. All of the data and theory used in this note have been in the public domain for many years[3].

The Model

We assume the simplest design for a first effort explosive, consisting of a solid Pu spherical core, very nearly a critical mass when surrounded by a high density tamping material, taken to be uranium. This larger sphere is then encased in the high explosive system which is designed to provide a converging spherical shock wave that would compress the assembly for a few microseconds before it flies apart. If the Pu core is nearly critical before the implosion, any compression will achieve supercriticality because a critical mass of fissile material depends inversely on the square of the density of the compressed material. For example, a compression of a factor 1.4 cuts the critical mass almost in half, so the core in this condition would contain ~2 critical masses. Introduction of neutrons into the core while it is in this supercritical state may start a

chain reaction culminating in an explosion. On the other hand, introduction of neutrons before maximum compression will result in less energy being released, which is a problem for those who want to use reactor grade Pu for explosives. The Pu obtained from uranium reactor fuel, will contain several Pu isotopes made by neutron capture or ejection because of the inescapable exposure of the created Pu-239 to the neutron flux in the reactor. The amounts of capture products, Pu-240, 241, etc. are dominated by the 1^{st} capture, Pu240; the next most abundant product, Pu-241, is so similar to Pu-239 that the two can be lumped together and considered to be a single fissile material for this purpose. Higher capture products will be ignored because significant production depends on long exposure, and we are interested in only moderately irradiated, "1^{st} cycle", LWR fuel. The real problem is that the Pu-240 nucleus has a small but finite probability of spontaneously breaking apart and releasing two or more neutrons as part of this process. The rate of this spontaneous fission is large enough to produce a sizeable neutron source in the amounts of fissile material we will consider.

The Chain Reaction

If a neutron is introduced into an assembly of fissionable material, it will end with one of several possible fates: 1) it can leak outside the material before it collides with a nucleus, or after making scattering collisions, 2) it may be captured by a nucleus with the emission of a gamma ray, 3) if it has enough energy it may knock another neutron out of a nucleus or 4) it may be captured by a nucleus which subsequently fissions and releases more neutrons. Taking all of these options into account, suppose that the average number of neutrons remaining after the first neutron disappears is k. Then, on average, each of these k will produce k more, or k^2, and so on. The total number is then $1+ k + k^2 + k^3 +$ etc $= (1-k)^{-1}$ for $k<1$. As k approaches 1 the total number becomes infinite, i.e. the chain continues indefinitely and the assembly is critical for the condition k=1. Of the four possible fates of each neutron listed above, the first two produce no new neutrons, and the third has such a small probability for neutrons of fission energy (too low) that it can be ignored, so the value of k is determined solely by the probability that a neutron causes a fission. If we denote the average number of neutrons per fission by ν and the probability that a neutron causes a fission by p_f, then, just at critical we can write $k=1=\nu p_f$

and $p_f = 1/\nu$ For Pu-239, the average number of neutrons emitted per fission is ~ 3, so that the probability that a neutron causes a fission at criticality, is just1/3; 2/3 leak out or are captured without causing a fission.

Just at critical, all the neutrons are required to maintain that state, including the very small fraction of delayed neutrons-those which appear seconds after fission rather just at the time of fission. The existence of these delayed neutrons allows the number of neutrons to be controlled by moving absorbing materials in reasonable times (seconds), and nuclear reactors are possible when operated at critical. Just above critical, (k>1), the neutron chain is maintained by prompt neutrons, and cannot be easily controlled. The time behavior of such a prompt chain is an exponential increase, the rate being determined by a quantity, α, which contains all the physics—the composition of the material, the change of the configuration with time, etc. The neutron population increases as $e^{\int \alpha dt}$, where $\int \alpha dt$ is called the number of generations—which tells us the number of neutrons and the amount of energy released by the fission chain after it has been initiated.

The value of α increases as the plutonium ball is compressed and then drops as it flies apart. If a chain does start, then when the number of generations reaches 43-45 or so, the pressure in the plutonium due to vaporization of the metal by the energy of fissions is equivalent to several kilograms of high explosive and will stop the implosion and begin the disassembly. If initiation occurs at nearly peak α, then more generations can occur during the time the device disassembles than if initiation occurs earlier at a lower value of α, because, of course, the generation time is just $1/\alpha$ If the chain produces ~ 56 generations,(i.e. another 11 to13 generations after the 43 to 45), approximately 20 kilotons of energy will have been generated.

Reactor Grade plutonium

As pointed out, the spontaneous fissions of the Pu-240 present in reactor irradiated uranium fuel provide a troublesome neutron source in the separated plutonium. Each gram of Pu-240 produces~1000 neutrons per second from this source[3], so that a ball of reactor grade plutonium weighing m kilograms and containing a fraction x of Pu-240 will produce $\sim 10^6$mx neutrons/second. Many of these neutrons would not cause fissions

or start a chain reaction—only 1/3 would at critical, as we have seen—they leak out or suffer non-fission capture. In fact, even though several neutrons are introduced at the same time, it is possible that none of them starts a chain reaction if the α and therefore the probability of fission is not large enough. At a fission probability of ~1/2, it can be shown[4] that the chance of starting an explosive neutron chain is ~1/3. To raise this probability from 1/3 to a sure fire 99% requires enough neutrons so that the chance of no chain is 1%; or mathematically, $(2/3)^n=.01$, where n is the number of neutrons needed. The solution of this equation is n=11.4 neutrons, and indeed, experience shows that about 10 neutrons are required for a 99% probability of actually starting a chain[3] in plutonium and we will use this number. We can then define a probability per second that a chain will be initiated in reactor grade plutonium as $s\sim10^5 mx$. But what we are really interested in is the probability; P, that the assembly will survive at least to an initiation time, t=T, that is expected to produce an explosion of energy yield Y. The time sequence of these events is: 1) the high explosive implosion starts to compress the Pu core, which becomes critical at time t=0, 2) at time t=T the neutron population in the now supercritical core becomes high enough to start a neutron chain reaction, either from unwanted spontaneous fissions, or by injection from an initiator, 3) about 45 generations later the device starts to disassemble due to the high pressures developed in the Pu, and 4), the remaining neutrons cause more fissions producing a total yield of Y kilotons as the material flies apart. It is easy to show that the survival probability, P, is related to the time, t, by the equation $P=e^{-st}$. If we now assume a linear connection between α and t, $\alpha=bt$, we can write $st=s\alpha/b$, where b, the rate of change of α, is related to the speed of compression, and can be taken as an indicator of the state of this aspect of the implosion art. Such a linear relationship between α and t is warranted if the compression is not too high, and greatly simplifies our numerical considerations. An important case is when initiation occurs at the earliest possible moment; t=T=0, when the core is just critical, then $\alpha=0$, and we obtain P=1 for the so-called fizzle yield, which is clearly a lower bound for the yield produced by the device. (P=1 means 100% probability that at least this yield will be attained.) This guaranteed yield was expected to be a little less than a kiloton for Trinity[2], and would be at least as large for the designs we are considering.

If the plutonium is substantially diluted with isotopes other than 239, then the change of reactivity must be taken into account in estimating the critical mass. We are particularly interested in fuel that is removed from a LWR after one cycle, which, on average contains ~83% Pu-239[5]. The 17% that is converted to other isotopes will be assumed to be about 14% Pu-240 and 3% Pu-241. The Pu-241 has, for this purpose, characteristics so close to Pu-239, that they may be lumped together, as mentioned above[2]. LANL has calculated that a bare critical mass of Pu-240 is ~40kg compared to16.28 kg for Pu containing 4.5% Pu-240[6]. A uranium tamped critical mass of Pu-240 can therefore be estimated (using the measured value of 5.91kg for uranium tamped 4.8% material and ignoring the difference from 4.5%) to be ~5.91 40/16.28=14.52kg, Using these numbers, the uranium tamped critical mass of plutonium of any fraction x of Pu-240 , assuming an approximately linear relation, is given by 5.50+9.02x kilograms. Inserting x=.14 in this formula, the critical mass of 1st cycle LWR plutonium, encased in a thick uranium shell, may therefore be estimated to be 6.73kg.

Probability Estimates

We will base the calculation of predetonation probability on data from the first actual experiment (the Trinity nuclear test) and the results of calculations provided by Carson Mark[2]. (It is interesting that these predictions were reported by Oppenheimer after the Trinity test, to apply to the Nagasaki bomb. Prior to Trinity, yield estimates by the prominent scientists were all over the map, but not for reasons of preinitiation, but rather from uncertainties about many aspects of the untested design[7]). To proceed, we need the composition and mass of the Trinity device. For this first test apparently very clean plutonium was obtained by frequent chemical processing of the uranium fuel at Hanford, resulting in a Pu-240 content of ~1%, and it is reported that data provided by Gen. Groves gave the mass as 6.2kg[8]. From the critical mass approximate formula in the previous paragraph, the critical mass of 1% material is just 5.6kg, less than the actual mass used for Trinity. This is because of a central void for an initiator, and somewhat different tamper. The LANL measured critical mass of 1% Pu with a central void for an initiator, presumably a mockup of Trinity, is 6.46kg[6].

We will take the above indicated changes into account as follows: we simply rely on the mathematical form of the probability, $P=e^{-st}$, where, since s is proportional to mx, a change to m`x` would imply that the new probability is $P`=P^{(m`x`/mx)}$. This assumes, of course, an implosion system of the same quality as that used for Trinity, but this too could be changed by simply reducing the time to the initiation value of α, T= α/b, by increasing b, the rate at which αincreases. Then the exponent of P would involve mx/b. The ratio b`/b can be increased without knowing the Trinity value, b, so we can speak of technical improvements in relative terms. These improvements would be in the high explosive system and the design of the core- tamper arrangement to achieve higher compression, faster, and in design of the core itself.[9]

. The values of the probability P are known only for yields of 20, 5, and 1 kilotons, because they are the only published values of P associated with the Trinity nuclear explosion: P=0.88, 0.94, and 0.98 respectively[2]. It is perhaps more perspicuous to present the results in terms of yield ranges. The results are presented in the following 3 charts in which the probability of achieving yields in the ranges ≤1kt(fizzle yield), 1 to 5kt, 5 to 20kt, and the nominal 20kt (design yield) is shown for the Pu-240 fractions considered. The published Trinity data have been added to the first chart since it applies to unimproved Trinity technology. Because of the rapid development of fission weapon technology in the late forties and early fifties we have characterized the charts corresponding to 2 and 3 times the Trinity implosion speed as “post Trinity” and “improved” technology. We do this for simplicity, even though the technological improvements may have been in several areas, such as new levitated pit and tamper design, new and more efficient initiators, and greatly improved high explosive design.

Conclusions

. The first chart shows that we can expect either 10% or 1st cycle (14%) LWR plutonium to provide a state operated laboratory with the basis for fission weapons with the most probable yield in the 1- 5 kilotons range, using only Trinity (1945) technology. Taking into account the wide availability of declassified nuclear weapon information and the enormous increases in computing and other technological aids since the Trinity shot

in 1945, it seems reasonable to attribute at least a doubling or tripling of the efficacy of the Trinity implosion system through the use of advances in implosion technology, initiators, and core design. The 6% Pu-240 material is included to correspond to current "weapon grade" plutonium. The 4.5% Pu-240 metal provides considerable improvement in reliability, but its production is of course a cost tradeoff. Critical masses of this grade of plutonium were reported[6] in the 1950's and correspond to a relaxation of the purity requirement post Trinity and pre boosting as implosion technique improved. The second and third charts are attempts to estimate the effect of such improvements on the expected yield. However, these estimates are based on unspecified extrapolations of the very first fission device design, by increasing implosion speed in the model, and are therefore rather crude estimates, but are expected to show the trend correctly and semi-quantitatively. Taking these caveats into account, chart 2 indicates that the 4.5% and 6% material will most likely achieve the design yield, and that the 10% material has some probability of being in the 5kt to 20kt range. The first cycle LWR 14% Pu has a flatter distribution and could produce a yield in the 1kt to 20kt range, but with higher yields having a higher probability. For the "Improved Technology" (Chart 3) all the Pu categories peak at the design yield, but the 14% data still leave the possibility of a somewhat reduced yield.

To take full advantage of the LWR material, boosting with some kind of fusion capsule would probably be required, but would likely need nuclear testing. Finally, however, it should be remembered that the fizzle yields of the devices considered will be at least as large as that of Trinity[2]; around 1kt, and that this guaranteed yield is already quite destructive.

CHARTS

All 3 charts show the probability of achieving yields in the ranges shown, for several Pu-240 fractions:

~1% was used in the Trinity test

4.5% corresponds to the Pu used during the years following Trinity before boosting was introduced in the 1950's,

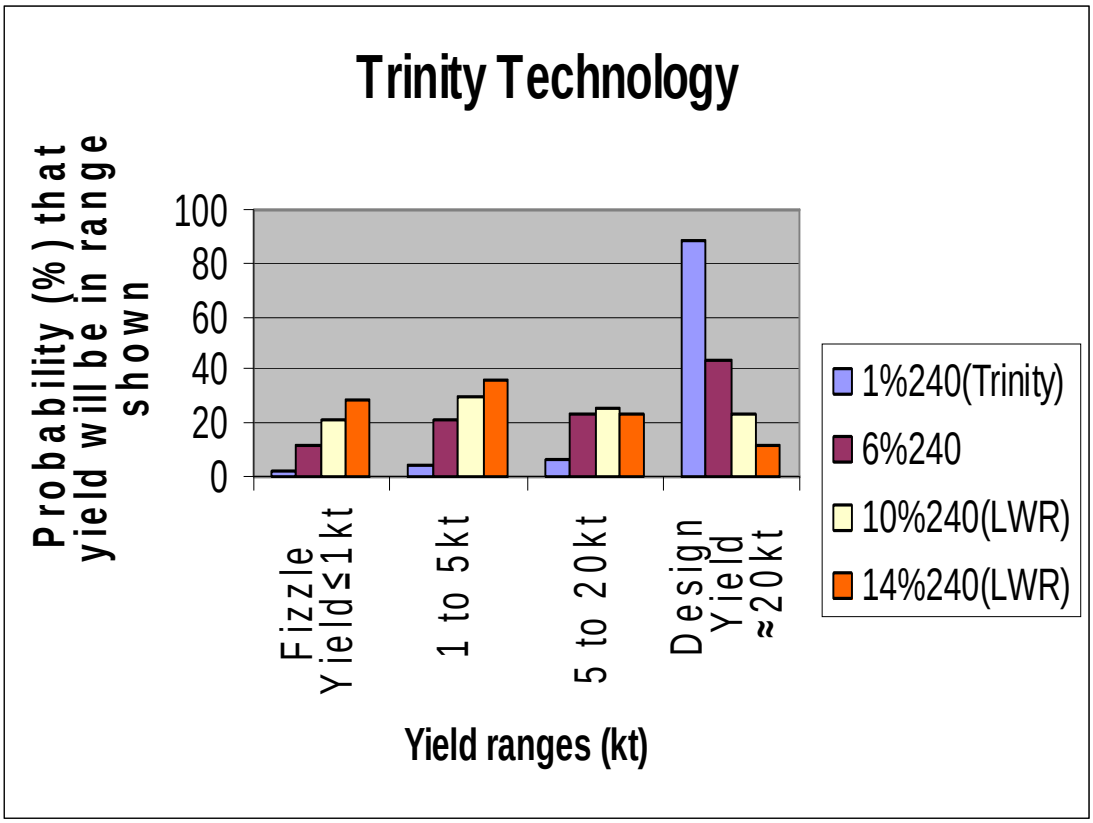
6% is a more or less standard "weapon grade" Pu,

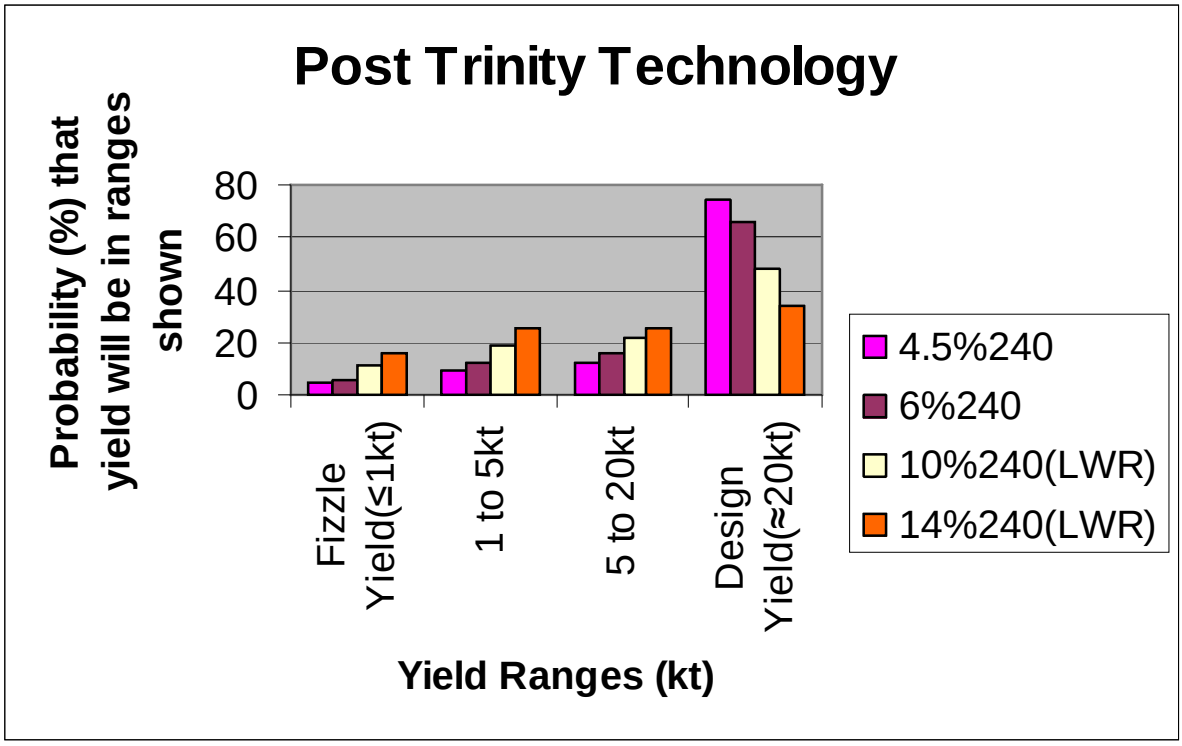
14% is the expected average 240 content of Pu withdrawn from a LWR after the first fuel cycle, and

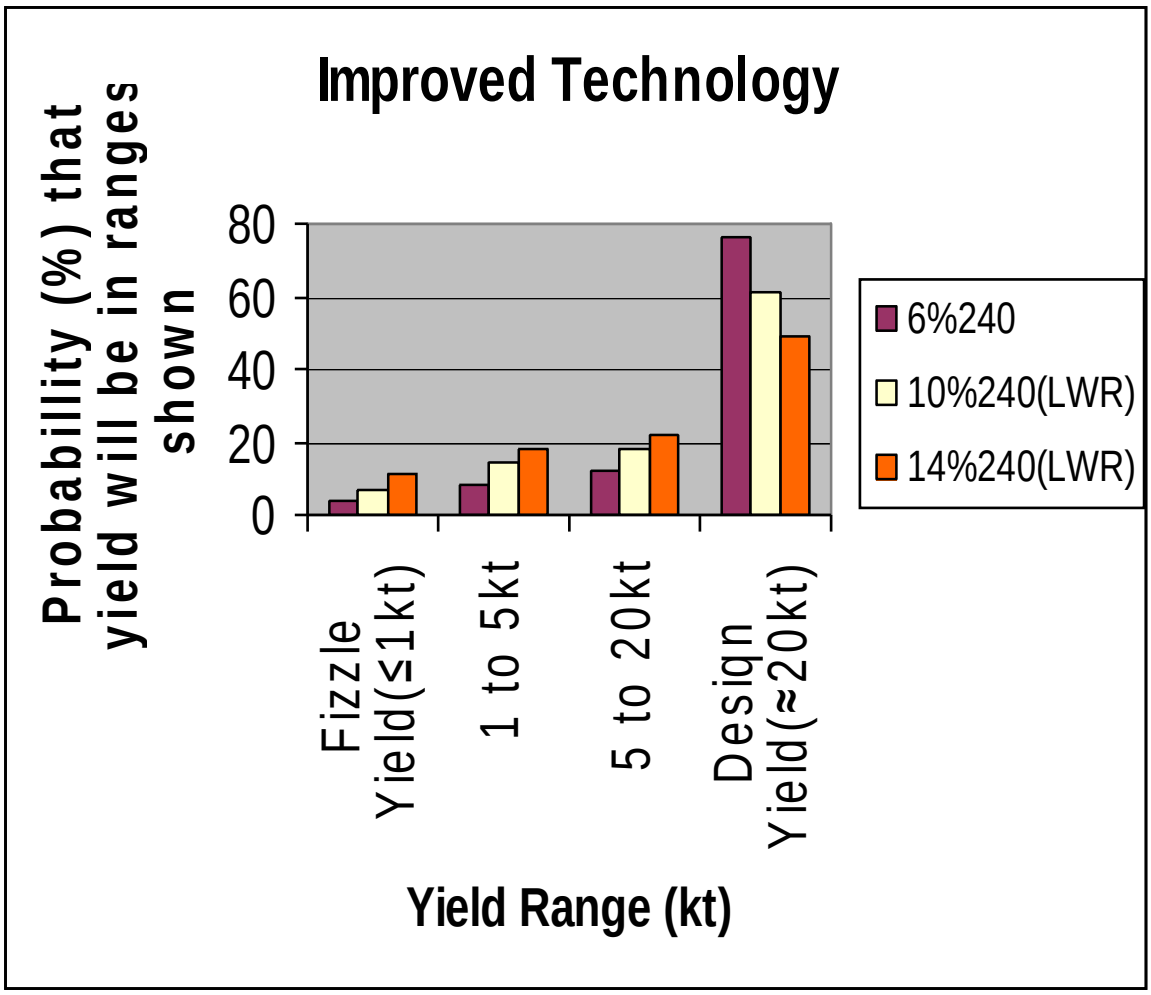
10% is withdrawn from the LWR somewhat earlier

The first chart applies to unimproved Trinity technology, and includes an addition of the published Trinity data for 1% Pu-240 plutonium as well as three Pu-240 fractions described above.

The second and third charts correspond, respectively, to 100% and 200% unspecified improvement in implosion technology over Trinity, which we believe can be considered to be the improvements made in the 10 years following the Trinity explosion. Given the vast changes in every pertinent aspect of technology and the availability of information on previous work since 1945, these assumed improvements are probably justified. The approximate nature of these correspondences should be kept in mind when using the numerical results; however, we believe that the charts present reasonable estimates of expected yield distributions.


Trinity Technology
Probability (%) that yield will be in range shown
100
80
60
40
20
0
Fizzle Yield≤1kt
1 to 5kt
5 to 20kt
Design Yield ≈20kt
Yield ranges (kt)
1%240(Trinity)
6%240
10%240(LWR)
14%240(LWR)


Post Trinity Technology
Probability (%) that yield will be in ranges shown
80
60
40
20
0
Fizzle Yield(≤1kt)
1 to 5kt
5 to 20kt
Design Yield(≈20kt)
Yield Ranges (kt)
4.5%240
6%240
10%240(LWR)
14%240(LWR)


Improved Technology
Probabillity (%) that yield will be in ranges shown
80
60
40
20
0
Fizzle Yield(≤1kt)
1 to 5kt
5 to 20kt
Design Yield(≈20kt)
Yield Range (kt)
6%240
10%240(LWR)
14%240(LWR)

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Notes

[1] "Non-Proliferation and Arms Control Assessment of Weapons-Usable Fissile Material Storage and Excess Plutonium Disposition Alternatives" U.S. Department of Energy. 1997 pp 37-39
Also "Management and Disposition of Excess Weapon Plutonium" The National Academy of Sciences 1994, pp 32,33.

[2]" Reactor Grade Plutonium's Explosive Properties"
J Carson Mark, Consultant, Nuclear Control Institute 1990

[3] For a discussion of the physics of nuclear explosives see "The Nuclear Weapons Complex: Management for Health, Safety, and the Environment" The National Academy of Sciences (1989) Appendix E "Physics of Nuclear Weapon Design"

[4] An approximate formula for the probability of chain initiation is $(\nu p_{f-1)} / \chi p_f$, where χ is the value of k(k-1)/2 averaged over the number distribution of neutrons per fission, where k is the number emitted , ranging from 0 to about 5. For Pu, $\chi \sim 3$.

[5]

[6] Plutonium critical masses are taken from H. C. Paxton, "Los Alamos Critical-Mass Data" LAMS-3067, 1964

[7] Rhodes, Richard, "The Making of the Atomic Bomb", Simon and Schuster, 1986

[8] Carey Sublette, "Nuclear Weapons Frequently Asked Questions" at http://gawain.membrane.com/hew/ This website contains a large amount of information on many aspects of nuclear weaponry in the public domain.

[9]To prepare the proper exponent, since only mass ratios are involved, and since Trinity was close to critical, and if we correct for both the central void and backing off from critical by correction factors, they are the same factors for any x, and will cancel out. Hence, for the pre-initiation calculation we can simply

use the solid ball critical masses, so that the proper exponent of P is (6.73 :1 4/5.59 :01) (b/b ')=16.85b/b '. For comparison, we make the same calculations for weapon grade Pu (6% Pu-240), and an intermediate, 4.5% Pu-240 core, for which a measured critical mass was published. The calculated critical mass from our formula is 6.04 kg and the exponent for the comparison with Trinity is 6.483 b/b ', for the 6% case, and 4.758b/b' for the 4.5% material, using the measured 5.91kg critical mass. This 4.5% material was apparently used in the period following Trinity and before the various advances that made this purity unnecessary