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Reactor-Grade Plutonium's Explosive Properties

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Fourth in a series of papers on issues bearing on extending and strengthening the Nuclear Non-Proliferation Treaty.

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These comments relate to the question of whether a terrorist organization or a threshold state could make use of plutonium recovered from light water reactor fuel to construct a nuclear explosive device having a significantly damaging yield. Three aspects of this question will be discussed separately:

I. Criticality Properties of Reactor-grade Plutonium;
II. Effects of Predetonation on Yield Distribution;
III. Some of the Problems Confronting a Terrorist Organization.

Finally, several conclusions are noted in IV.

This paper is being written at this time because questions appear to persist in some non-proliferation policy circles about whether a bomb really could be made from reactor-grade plutonium of especially high burnup and whether the task is too daunting for a threshold state or terrorist group, even if it is technically feasible.

This paper is derived from information in the public domain. It is appropriate, therefore, to make the information and its significance available to policymakers and members of the public who are concerned about preventing the spread of nuclear explosives.

The question of whether terrorists could build nuclear weapons was also examined by this author and four colleagues in a paper prepared in 1986 for a task force on nuclear terrorism.[1]

I. Criticality Properties of Reactor-grade Plutonium

The original implosion assembly system used in the Trinity test in 1945 was capable of obtaining 20 kilotons from weapons-grade plutonium. In the weapons tests conducted in 1948 it was shown that an assembly system of the same size could also handle U-235 effectively. From the discussion below it may be seen that such an assembly system would be capable of bringing reactor-grade plutonium of any degree of burnup to a state in which it could provide yields in the multi-kiloton range. The original implosion system had a diameter of less than 5 feet, including an outer aerodynamic case. Thus, it does not (as was recently suggested) require a "device of the dimensions of a fair sized room" to handle reactor grade plutonium. Moreover, it is well known that the design of the first implosion system was quite conservative, and that there are a number of straightforward improvements which could be implemented to reduce the size of the device on the basis of laboratory-type experiments without having to resort to nuclear tests.

Discussion

In addition to the isotope Pu-239 the plutonium extracted from spent LWR fuel may contain appreciable fractions of other plutonium isotopes formed as a result of successive neutron capture or n-2n reactions. At very low burnup levels the fractional amounts of the secondary isotopes are very small. At a level of a few thousand megawatt-days per metric ton (MWD/MT), for example, the fraction of Pu-240 may be a few percent of the total plutonium, with the fraction of Pu-241 being approximately an order of magnitude smaller, and that of Pu-242 an order of magnitude smaller still. At higher burnups these fractional amounts increase so that at a very high level (≈50,000 MWD/MT or so—about as high as current interest appears to extend) a pattern of the following general sort could be approached:

(Pu-239: Pu-240: Pu-241: Pu-242) = (.40: .30: .15: .15).

Other plutonium isotopes would also be present, but in relatively small amounts. The most prominent of these would be Pu-238, which could reach a level of a few percent in very high burnup material. This would not have a significant effect on critical masses. But because of its relatively short half-lives for alpha decay and spontaneous fission, the amount of Pu-238 might need to be taken into account in determining the alpha activity or neutron source in plutonium from highly exposed reactor fuel.

Each of the plutonium isotopes is sufficiently fissionable that the separated isotope in metal form could provide a bare critical mass, so that a bare critical assembly could be made with plutonium metal no matter what its isotopic composition might be. The odd isotopes (Pu-239 and 241) are both "fissile"—that is, fission may be induced in them by neutrons of any energy, whether slow or fast. Their fission cross sections differ in detail but are similar enough that their bare critical masses are nearly equal, being about 15 kg in δ-phase metal (ρ = 15.6 g/cc). The isotope Pu-238 is "fissionable"—that is, only neutrons with energy above some threshold can induce fission. However, the Pu-238 threshold is at some quite low energy and its fission cross section above about 0.5 MeV is larger than that of Pu-239. In spite of producing fewer neutrons per fission (2.75 vs. 3.0) the bare critical mass of Pu-238 in δ-phase metal is also ≈15 kg.

. . . [a Trinity-type device] would be capable of bringing reactor-grade plutonium of any degree of burn-up to a state in which it could provide yields in the multi-ton range.

For Pu-240 the fission threshold is at a few hundred kilovolts; but above 1 MeV the fission cross section, though smaller than that for Pu-239, is larger than that for U-235. The number of neutrons per fission (≈3) is the same as that for Pu-239, 241, and 242. The bare critical mass of Pu-240 in δ-phase metal is about 40 kg. This is smaller than that for 94% U-235 in uranium metal at normal density (ρ = 18.7 g/cc), which is ≈52 kg. Thus Pu-240 is a significantly more effective fissionable material than 94% U-235 in a metal system. It should be noted, however, that this relative superiority would not carry over to the same extent for these materials in the form of oxides. In PuO_2 or U-235 O_2 the average energy of the neutrons is reduced appreciably by their scattering on oxygen. In a Pu-240 O_2 system, therefore, some fraction of the neutrons in the spectrum applicable to a metal system will be moved to energies near or below the Pu-240 threshold where the Pu-240 fission cross section is poor, whereas the fission cross section of U-235 holds up for such lower neutron energies.

At energies above 1 MeV, the fission cross section of the isotope Pu-242 is quite similar to that of Pu-240, but it is a less effective fissionable material because its fission threshold is about a hundred keV higher. The bare critical mass of Pu-242 in δ-phase metal has been calculated to be ≈177 kg. To bring this more in line with the other isotopes, one can think of replacing the Pu-242 component with a new component consisting of a 50-50 mixture of Pu-242 and Pu-241,

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with the material taken from the Pu-241 fraction since there is enough of that to supply what's necessary even at the extreme high burnup level considered. The fission cross section of this new component, which is the average of the cross sections of Pu-241 and Pu-242, is quite similar to that of Pu-240 in the range of energies above 1 MeV, and considerably larger at lower energies where the Pu-240 and 242 cross sections fall away, while that for Pu-241 does not. Thus, the material of the new component is superior to Pu-240, which in turn is superior to U-235. At all burnup levels, then, the critical mass of reactor-grade plutonium is intermediate between that of Pu-239 and U-235.

By the use of a reflector a few inches thick, the critical mass of all these materials can be reduced by a factor of two, or so, below the bare critical mass; and, at least provided the reflector is of some heavy metal so as not to moderate the neutrons to an important extent, the relative ranking of the critical masses will be preserved.

II. Effects of Predetonation on Yield Distribution

One week after the first fission explosion on July 16, 1945, Robert Oppenheimer wrote to General Leslie Groves' deputy and described the expectations concerning the use of the Trinity device in combat.[2] He said: ". . . The possibility that the first combat plutonium Fat Man will give a less than optimal performance is about 12 per cent. There is about a 6 per cent chance that the energy release will be under 5000 tons, and about a 2 per cent chance that it will be under 1000 tons. It should not be much less than 1000 tons unless there is an actual malfunctioning of some of the components." One week later General Groves wrote to the Chief of Staff: "There is a definite possibility, 12 per cent rising to 20 per cent, as we increase our rate of production at the Hanford Engineer Works, with the type of weapon tested that the blast will be smaller due to detonation in advance of the optimum time. But in any event, the explosion should be on the order of thousands of tons."

Evidently both Oppenheimer and Groves were referring to what will be identified in the following discussion as the "fizzle yield"; that is, the smallest nuclear yield this particular device would provide. They do not state a value for this yield; but in view of their saying "it should not be much less than 1000 tons" it may be presumed that they were thinking of some value like 700 tons, or so. The effect of using reactor-grade plutonium in this assembly instead of the high purity plutonium used in 1945 would be to increase the probability that the yield realized would fall short of the levels mentioned by Oppenheimer, but it would not greatly change the actual value of the fizzle yield—which would always be equaled, or exceeded.

In the following discussion some indication is given of the differences between plutonium and highly enriched uranium with respect to pre-detonation and fizzle yields.

Discussion

In any supercritical system, the number of neutrons, the rate of fission, and the level of energy generated increase exponentially—that is, they all vary with time in a way which may be written as $e^{\alpha t}$. The value of the time constant α, which is zero in a system which is just critical and in which the neutron population remains constant, may be as large as one, or a few, times 10^8/sec in a highly supercritical metal system of U-235 or Pu-239. Obviously, the value of α increases with the degree of supercriticality (since a smaller fraction of the neutrons escape without causing a fission), with the density of the fissile material (since, with the atoms closer together, the distance and time for a neutron to cause a fission is reduced), with the average neutron velocity (which is higher in metal than in oxide, for example), and with the factors which favor small critical masses.

Independent of the value of alpha, nothing of much consequence occurs in a supercritical system containing only fissile material in the core until the energy level becomes high enough to vaporize all that material. Only then do pressures build up which can force a disassembly or halt the motion which may be driving the assembly towards a more supercritical condition. At about that point the core begins to expand, and its density starts to drop, and the value of alpha (as also the degree of super-criticality, and the rate of increase of the neutron population and energy generation) begin to decrease rapidly toward zero (at which point the system is critical and the neutron population and the energy generation rate are at, or near, their maximum) and on to negative values (where the neutrons rapidly leak away, the energy generation rate falls off, and the reaction is over). Typically, most of the energy from the reaction is developed during this disassembly phase.

. . . a bare critical assembly could be made with plutonium metal no matter what its isotopic composition might be.

As indicated by Robert Serber in the "Los Alamos Primer" of April 1943[3], on the basis of an approximate calculation valid only for a small degree of supercriticality, in any particular system the efficiency of the reaction (the fraction of the fissile material actually consumed) will be proportional to the third power of alpha at the time the motion of disassembly first gets well under way. In a core with a mass of 10 kg, or so, this stage will be reached when the value of $\alpha \cdot t$ is somewhere between 40 and 45, where t is measured from the time the chain reaction is initiated. If the system is highly supercritical when the chain starts, so that $\alpha \approx 10^8$/sec, say, then the time for $\alpha \cdot t$ to reach a value ≈ 45

will be extremely short, and there could be rather little change in the degree of supercriticality (or alpha) during this time. However, had the chain started much earlier in the assembly process, when the value of alpha was much smaller, there could have been an appreciable change in alpha during this incubation period—while the 45 generations (as they might be called) were being accumulated. In such a case one would consider the $\int\alpha \cdot dt$ (rather than $\alpha \cdot t$) taken from the time the chain started till $\int\alpha \cdot dt = 45$, and the explosion alpha would be the value applying at the end of that period. Clearly, the smallest possible explosion alpha will be that resulting from a chain which started just as the system reached critical (and alpha reached zero) in the course of its assembly. The yield resulting from this situation will be the smallest possible, and has been referred to as the "fizzle" yield.

Oppenheimer's breakdown of probabilities may be rephrased in the following way, namely: that, with the implosion assembly system and the high grade of plutonium being used, the probability was 0.88 that a device would survive long enough without a chain being initiated that it would provide the nominal yield; about 0.94 that it would survive long enough that the yield would be greater than 5 kilotons (one quarter of the nominal); about 0.98 that it would survive long enough to provide a yield in excess of one kiloton. Only in 0.02 of all firings would a chain be initiated so early that the energy release would be between the fizzle yield and one kiloton. Were one to change only the strength of the neutron source (which arises from spontaneous fission and alpha-n reactions) while keeping the mass and reactivity of the fissile material and everything else the same, these probabilities would change. Were the neutron source twice as large, for example, the probability of realizing the nominal yield would be only $(0.88)^2$, *u.s.w.* In particular, for sources 10, 20, 30, and 40 times larger than the one which applied at Trinity these probabilities (and the fraction initiated very close to critical) would be as shown in the following Table.

	Yield: Nominal	above 5 kt	above 1 kt	Fizzle to 1 kt
Source:				
Trinity	.88	.94	.98	.02
10 X "	.28	.54	.82	.18
20 X "	.08	.29	.67	.33
30 X "	.02	.16	.55	.45
40 X "	.006	.08	.45	.55

The largest of the sources above is most probably larger than that in the most heavily exposed plutonium considered earlier. It will be seen that as the neutron source is increased from a low level to a very high level the distribution of yields realized changes from one in which the nominal yield is the typical yield and very severe predetonation is rare, to one in which the nominal yield is rare (though never completely excluded) and the typical yields are in a band from one to a few times larger than the fizzle yield.

With the improved data and greatly improved calculation capability which have become available in the meantime, the particular values quoted by Oppenheimer in 1945 would no doubt require some revision. The substitution of a somewhat larger mass of reactor-grade plutonium for the high-grade plutonium employed in the Trinity device would also lead to some changes, both in the nominal yield and the fizzle yield. However, the general pattern pictured above would continue to apply: in the same assembly system some mass of reactor plutonium of any grade would (since this asssembly system was capable of making effective use of U-235, which is a less reactive material than reactor-grade plutonium) have a nominal yield of ≈10kT or more, and an associated fizzle yield of a few percent of its nominal yield—which is to say, some hundreds of tons. Under heavy predetonation the yields realized would most frequently fall in the range of one to a few times the fizzle yield—never less, but occasionally many times larger. Though almost all of these yields are much smaller than the nominal yield, they would nevertheless constitute quite damaging explosions, and are not reasonably dismissed as "duds" as has sometimes been suggested.

By the use of a reflector a few inches thick, the critical mass of all these materials can be reduced by a factor of two, or so, below the bare critical mass . . . .

As a final comment concerning fizzle yields it may be noted that the more rapidly the criticality (or alpha) of the fissile material increases after it first becomes critical the larger the value of alpha at the moment when $\int\alpha \cdot dt = 45$. If, for example, we assume that alpha increases linearly with time, so that $\alpha = k \cdot t$, then, when $\int\alpha \cdot dt = 45$, we have $t = \sqrt{(90/k)}$ and $\alpha = \sqrt{(90 \cdot k)}$—which is larger, the larger k may be. Since the efficiency of the fizzle explosion varies as the cube of this value of alpha, the faster the assembly proceeds the larger the fizzle yield of a given mass of fissile material. From the fact that the Trinity assembly was a very conservative design, it would seem likely that straightforward ways could be found to realize a faster-moving implosion, which could have the effect of increasing fizzle yields to higher levels than those applying above.

On the other hand, since the time interval from first critical to complete assembly might be something like 50 times longer in a gun-assembly system than in an implosion —so that the slope of the alpha-curve (the value of k, above) would be much smaller—not only would initiation be essentially guaranteed early in the assembly process even by the neutron source in very high-grade plutonium, but the value of alpha at the earliest possible explosion time would be smaller by a factor of something like $\sqrt{50}$, and the fizzle yield would be reduced by a large factor. Thus, not even the best weapons-grade plutonium is of any interest in connection with a gun-type assembly system.

The effect of using reactor-grade plutonium . . . would not greatly change the actual value of the fizzle yield—which would always be equaled or exceeded.

These considerations come out quite differently in connection with highly enriched uranium because the neutron source from spontaneous fission in such material is smaller than that in even the best grades of plutonium by a factor of more than a thousand. In the relatively slow-moving gun-type device one might wish to assemble a couple of critical masses, or so, which would imply bringing together something like 50 kg of 94% U-235, since the critical mass with a reflector can be about half the bare critical mass of 52 kg. The fizzle yield of such a system would, again, be some uninteresting low value; but, with the very low neutron source which could be realized in this material, the probability of initiating a chain at a very early stage of the assembly process may be small enough to ignore. Indeed, Luis Alvarez, a scientist with the Manhattan Project during its war years, has said,[4] "With modern weapons-grade uranium the background neutron rate is so low that terrorists, if they had such material, would have a good chance of setting off a high-yield explosion simply by dropping one half of the material on to the other half." What he meant by "high-yield" or "good chance" are not explained; but his mere statement calls attention to the fact that highly enriched uranium is in a class by itself.

III. Some of the Problems Confronting a Terrorist Organization

• **Technical Personnel.**

Competence and thorough understanding will be required in a wide range of technical specialties. These include: shock hydrodynamics, critical assemblies, chemistry, metallurgy, machining, electrical circuits, explosives, health physics, and others. At least several people who can work as a team will be needed. These will have to be carefully selected to ensure that all necessary skills are covered, but they need not have been previously engaged in designing or building nuclear weapons.

• **Costs.**

In addition to support for the personnel over a period adequate for planning, preparation and execution, a considerable variety of specialized equipment and instrumentation will be required, all or most of which can be obtained through commercial sources.

• **Hazards.**

Radiation, criticality, the handling of noxious materials and explosives all present potential hazards which will have to be foreseen and provided against.

• **Detection.**

Assuming the operation is contrary to the wishes of the local national authorities the organization must exercise all necessary precautions to avoid detection of their activities. They would no doubt be faced by a massive search operation employing the most sensitive detection equipment available once it should be known that someone had acquired a supply of material suitable for use as an explosive.

• **Acquisition.**

Very early in its planning and equipment procurement phase the organization will need information concerning the physical form and chemical state of the fissile material it will have to work with. This will be necessary before they can decide just what equipment they will need. The isotopic content of the material could be determined by straightforward means. The actual acquisition of the material would probably be the responsibility of a separate task force for which the problems and hazards would be those set by the safeguards and security authorities.

IV. Conclusions

1. Taking "weapon" to signify an object suitable for stockpile by a military organization, then heavily irradiated reactor plutonium would not be attractive for an arsenal of pure fission devices. For that purpose one would wish to have a set of warheads with a reliable known yield. One would also wish to have objects which could be turned out in a production-line fashion. However, for a terrorist organization acting alone or on behalf of a rogue state, with interests focused on the possible use of one, or a very few, devices, these considerations might be weighed quite differently. In addition, radiation exposures associated with fabrication which might be unacceptable for a sustained activity might not be troublesome for a one-shot operation.

2. It has been suggested that the fact that the U.S. appears to have made only one experiment using reactor-grade plutonium and has not chosen to adopt it for regular weapons production indicates that such material is of little worth. That is not the correct interpretation. There is, of course, no question but that weapons-grade material is preferable from a design standpoint; and if, as for the U.S., one has the option and is paying for the plutonium anyway, one chooses the most advantageous. So would the terrorist if he had a choice. But if he can't get weapons-grade material he would take whatever he can get, should any be open to him.

3. The technical problems confronting a terrorist organization considering the use of reactor-grade plutonium are not different in kind from those involved in using weapons-grade plutonium, but only in degree. For example, it is of great importance to avoid the inhalation of plutonium dust or vapor; but the provisions which would be adequate for weapons- grade material would require little, if any, modification to be acceptable for reactor-grade material. The hazards and difficulties associated with assembling a device would be less if highly enriched uranium were used.

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4. The method of coping with the problems and difficulties of making an explosive device with reactor-grade plutonium is entirely in the hands of the terrorist organization. The information necessary to meet the needs is available, and can be assembled by a properly chosen team of specialists. It cannot be said whether or not they would conclude that the effort involved is within their reach, or "worthwhile", since that depends on many factors known only to them. It can be said that the only point on which established authorities can influence their decision is on that of the acquisition of material. Whether that should be more or less difficult, and whether or not the fact of their successful acquisition would be known rapidly and with assurance, could be important in this respect.

5. Assuming they do not also have access to a supply of highly enriched uranium, and assuming that the working group in question has been specifically formed to produce a first device in as short a time as possible with a high degree of confidence in obtaining a significant nuclear yield, the amount of material they would have to acquire could scarcely be as small as 5 kg, though it might not have to be very much larger than 10 kg. Even for a working group with the time and the means of conducting an extended series of non-nuclear assembly experiments—circumstances more likely to apply to a group engaged in a national effort by some Nth country than to a terrorist group—an amount of at least several kilograms would be necessary.

6. It has been suggested that rather than trying for an explosive device a terrorist organization might merely set out to disperse a quantity of reactor-grade plutonium in some highly populated location. This would bypass many of the difficult technical problems involved in producing an explosive device; and in this case reactor-grade plutonium, being several times more noxious than weapons-grade, could be the material of choice. However, it is not fully clear what objective would be realized by actually going through with such an action which could not be met as well, or better, by a well-publicized and credible threat. Here, again, the main line of defense available to the authorities is to ensure that the acquisition of such material is difficult, and that they have the means of assuring themselves rapidly whether or not the material claimed to be available is missing.

7. Finally, if methods of separating plutonium isotopes using laser technology (already receiving serious consideration in the U.S.) should, in the future, come within the reach of many industrial states, then stocks of reactor-grade plutonium would present a much more direct access to proliferation of nuclear weapons than they may appear to do at present.

ENDNOTES

1. J. Carson Mark, Theodore Taylor, Eugene Eyster, William Maraman and Jacob Wechsler, "Can Terrorists Build Nuclear Weapons?", in *Preventing Nuclear Terrorism: The Report and Papers of the International Task Force on Prevention of Nuclear Terrorism*, Leventhal and Alexander, eds., Lexington Books, 1987, pp. 55-65.

2. Quoted by Albert Wohlstetter in *Foreign Policy*; 25, winter, 1976-77; p. 160.

3. R. Serber, "The Los Alamos Primer", Report L.A. 1, April, 1943. (Declassified in 1965.)

4. Luis W. Alvarez, *Adventures of a Physicist*, Basic Books, New York, 1987, p. 125.