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Proliferation-Resistant Nuclear Fuel Cycles

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PROLIFERATION-RESISTANT NUCLEAR FUEL CYCLES

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ABSTRACT

The properties of plutonium containing unusually large proportions of the ^{238}Pu isotope are considered in relation to resistance to nuclear proliferation. Several fuel cycle modifications for light-water reactors are evaluated. It is shown that the ^{238}Pu isotopic concentration can be increased substantially from the approximately 1.5% in discharged fuel from reactors operating presently. Concentrations of 8 to 10% are readily achievable, and values approaching 20% may be practical. The increased ^{238}Pu content is accomplished by increasing its production by recycling ^{236}U (via recycle of uranium from spent fuel) and ^{237}Np, and by decreasing production of isotopes heavier than mass 238 by substituting thorium for ^{238}U. Impact on the fuel cycle appears to be no more severe than that from other proliferation-resistant concepts currently under consideration. The properties of such plutonium, particularly heat generation, may offer a viable deterrent to weapons use of the material by organizations without appropriate experience and technology, thereby substantially reducing the risk of nuclear proliferation.

INTRODUCTION

Early in 1977 several Union Carbide staff members independently began considering the possibility of a nuclear fuel cycle that would produce plutonium having a high deterrence to nuclear weapons use.[1,2] This report is an attempt to summarize and record the information that has been developed.

Fissile uranium isotopes can be "denatured" by diluting them with ^{238}U, thereby increasing the critical mass beyond practicality. Since all plutonium isotopes have reasonably small critical masses, this concept is not applicable to plutonium. Any deterrent effect in this case must result from properties other than criticality, but criticality has previously been emphasized to the virtual exclusion of any other factor.

The essential steps in the utilization of a potential weapons material exist in the recovery and fabrication of plutonium, and in the assembly, storage, and delivery of a nuclear device. Any physical property that can be enhanced to such a degree that these steps are made more

difficult will add some degree of deterrence toward weapons application.

There are several properties of the isotopes of plutonium that, if sufficiently enhanced, produce a composite plutonium mixture which may be a credible deterrent. These properties of (1) heat generation, (2) neutron production, and (3) radiation intensity are all naturally increased as methods are employed to increase the ^{238}Pu content in the plutonium isotopic mixture.

The concept proposed here is that a ^{238}Pu "spike," if it can be produced in sufficient magnitude, may provide a credible, non-chemically-removable deterrent to the use of power reactor plutonium as a weapons material. It will be shown that reasonable modifications of the fuel cycle can result in production of plutonium containing some ten times more ^{238}Pu than in the case of the present once-through LWR cycle.

It is most significant that, if this concept is viable, a truly denatured fuel cycle can be achieved only if fuel is reprocessed. The denaturing results from the recovery and recycle of uranium and neptunium. In this context, LWR fuel reprocessing would need to be initiated to obtain neptunium and recovered uranium containing ^{236}U for use in denaturing plutonium produced in future LWRs, and, at the same time, produce a denatured plutonium stockpile high in ^{238}Pu for use in future LMFBRs.

This approach is not an absolute deterrent to the diversion of power reactor-generated plutonium to weapons use, and it has always been clear that there is no practical absolute deterrent. However, various steps can be taken to make plutonium less attractive for weapons use. One might suggest that the problem is "solved" when a weapons production route independent of power reactors is more attractive than diversion of power reactor fuel. All such considerations are clearly subjective. It is our hope that this proposal can be evaluated on the same basis as other alternatives, such as the various thorium reactor fuel cycles,[3,4] recycle schemes (CIVEX),[5] improved physical security measures, or the outright exclusion of nuclear energy as a civilian power source.

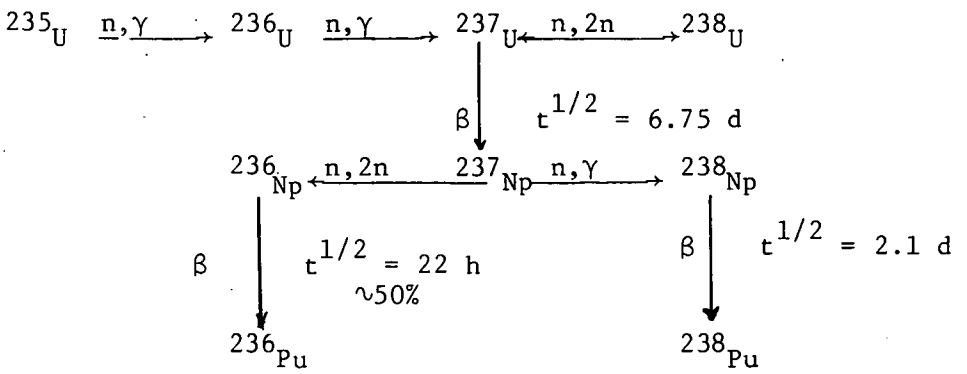
The analyses that have been done in support of the use of ^{238}Pu as a denaturant have covered the following four general areas:

1. methods for increasing the ^{238}Pu content in power reactor plutonium,
2. estimates of potential heating problems,
3. radiation hazards from neutron and gamma emissions, and

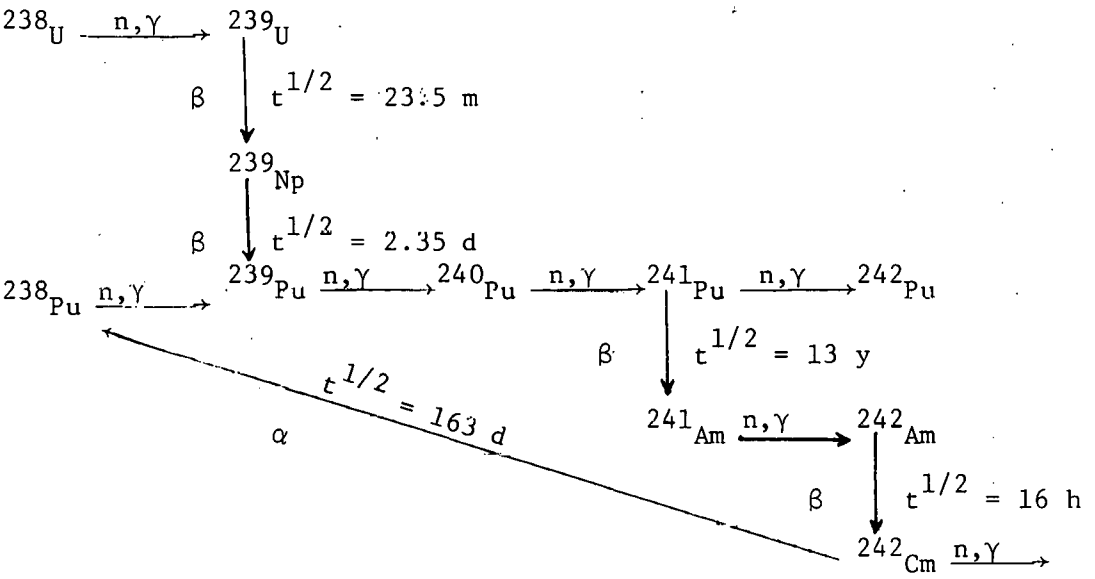
4. resulting complications to the power reactor nuclear fuel cycle.

^{238}Pu PRODUCTION METHODS

Plutonium-238 is produced in nuclear reactors principally through two routes, from ^{236}U neutron capture and (to a smaller extent) from ^{238}U (n,2n) reactions, as shown below:



A secondary route, which will be of importance if plutonium recycle is utilized, results from the decay of ^{242}Cm. This route is outlined below:



With the standard PWR once-through fuel cycle, having an initial ^{235}U enrichment of 3.2 wt %, the discharged fuel contains about 10 g of plutonium per kilogram of total fuel. The ^{238}Pu concentration in the plutonium is about 1.5 wt %. From considering the production routes, it is apparent that the ^{238}Pu fraction in the total plutonium can be enhanced if any or all of the following approaches are employed:

1. Recover and recycle the neptunium, and perhaps americium and curium isotopes (in existing flowsheets, these are routed to waste).
2. Directly recycle uranium in which the ^{236}U content is significant (present plans are to recycle uranium via enrichment, during which, under expected operating conditions, a substantial fraction of the ^{236}U is lost to tails).
3. Reduce the ^{238}U fraction in the power reactor fuel by substitution of thorium as the fertile material (this is the thorium-denatured uranium fuel cycle that has been proposed for other reasons).

Items (1) and (2) enhance ^{238}Pu production, while (3) reduces the production of isotopes heavier than ^{238}Pu.

Several ways of modifying the standard fuel cycle have been considered in an effort to increase significantly the ^{238}Pu content of the resulting plutonium. Some of these are described in Table 1. The resulting ^{238}Pu concentrations from each fuel cycle are shown in Fig. 1. It is apparent from this figure that the ^{238}Pu concentration can be significantly increased by the choice of fuel cycle. Case 1 shows the effect of continued recycle of the ^{236}U and ^{237}Np on the ^{238}Pu concentration in the uranium fuel cycle. Cases 2 and 5 show the additional effect of substituting thorium for some of the ^{238}U, which also decreases the total plutonium production. Case 3 shows that a PWR fueled with denatured thorium breeder blanket uranium produces plutonium having much lower ^{238}Pu concentrations. Case 4 shows that, after one recycle with the thorium-^{235}U denatured cycles, the ^{237}Np may be withdrawn (at some reduction in proliferation resistance because of the ^{235}U like weapons potential of the neptunium) and used to denature other reactors, and the resulting plutonium from successive cycles will still have a high ^{238}Pu concentration.

Table 1. PWR fuel cycle variations to increase ^{238}Pu concentration[a]

1.	Complete recycle of recovered uranium, plutonium, and ^{237}Np from standard uranium cycle. Fissile makeup with 20% ^{235}U. The first cycle (the left end of the curve in Fig. 1) represents the once-through or stowaway cycle presently in effect, with a ^{238}Pu concentration of 1.47%.
2.	Thorium-20% ^{235}U denatured cycle, with recycle of all uranium, plutonium, and ^{237}Np. Fissile makeup with 20% ^{235}U.
3.	Thorium with fissile uranium from thorium breeder blanket (^{233}U) denatured with diffusion plant tails. Recycle of all uranium, plutonium, and ^{237}Np. Fissile makeup from thorium breeder blanket uranium.
4.	Thorium-20% ^{235}U denatured cycle, with recycle of all uranium, plutonium, and ^{237}Np, until the ^{238}Pu content is $\geq$ 7% in discharge plutonium; then only uranium and plutonium are recycled. Fissile makeup with 20% ^{235}U. Neptunium-237 may be recovered, under proliferation resistant conditions, for use in other reactors.
5.	Thorium-20% ^{235}U denatured cycle, with recycle of only uranium and ^{237}Np. Fissile makeup with 20% ^{235}U. Recovered plutonium may be used as denatured breeder core fuel.

[a]These five scenarios are compared in Fig. 1.

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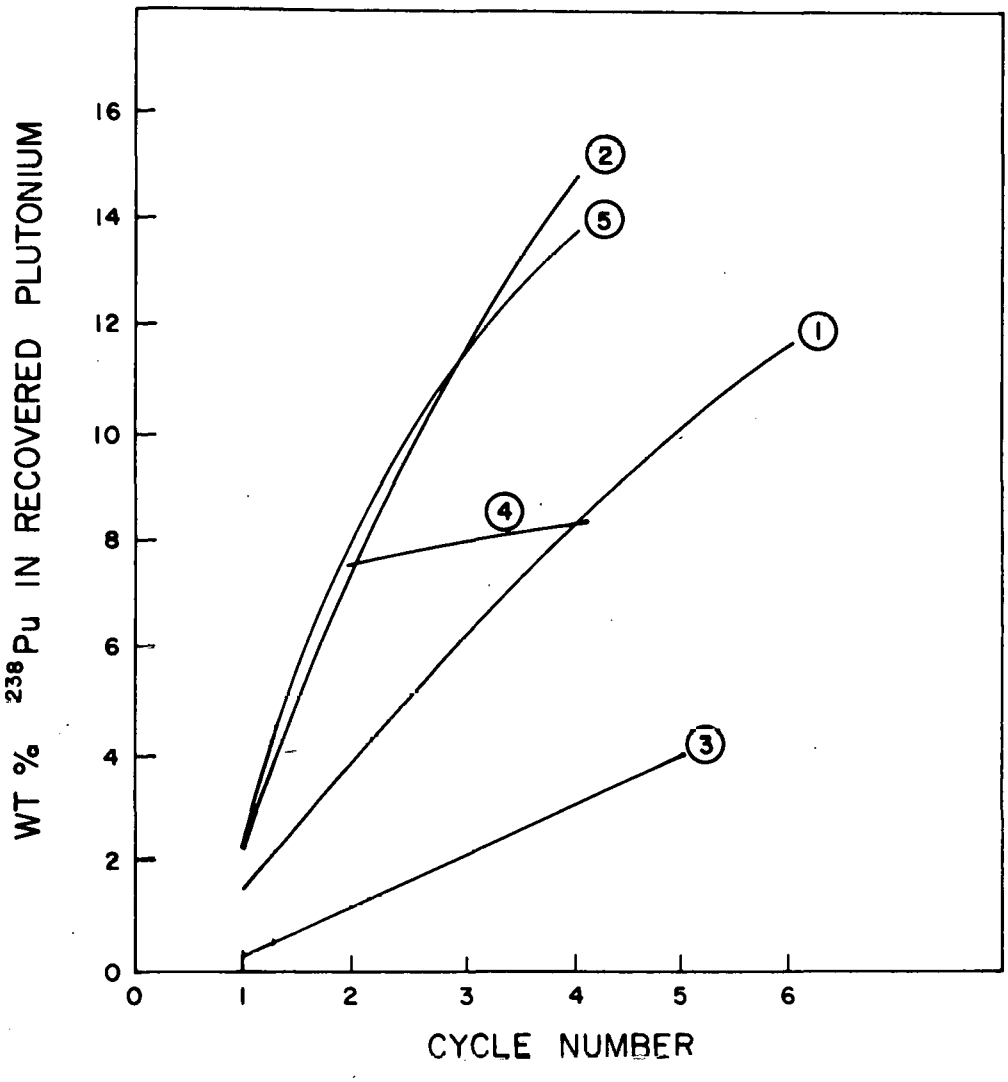

Fig. 1. Concentration of ^{238}Pu in PWR discharge plutonium for several fueling schemes.

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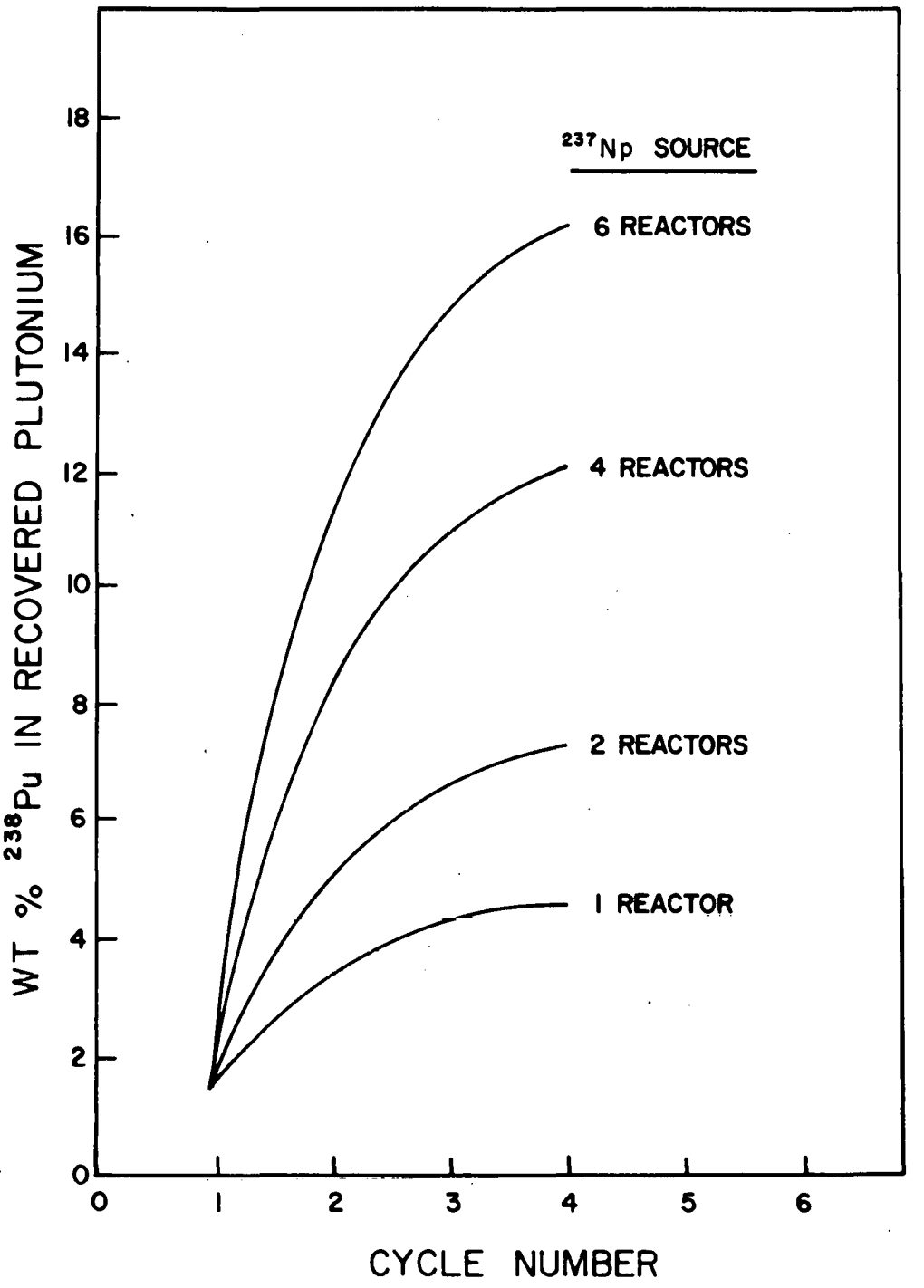

Fig. 2. Concentration of ^{238}Pu as a function of ^{237}Np contributors and number of recycles.

One could use excess ^{237}Np by adding it to the fresh, slightly enriched uranium fuel of existing or new PWRs, so that the plutonium produced would have a higher ^{238}Pu content. This effect, which is illustrated in Fig. 2, was analyzed by assuming that the ^{237}Np from a standard first-cycle PWR could be recovered and added to fresh fuel for other similar reactors. If the ^{237}Np from one reactor is recycled only to itself or another single reactor, the resulting ^{238}Pu concentration is represented by the lowest curve. If the ^{237}Np from two reactors is recycled to one reactor, the second curve results. Thus, even in this simple recycling scheme, high ^{238}Pu concentrations are quickly attainable in a fraction (approaching half) of all the reactors.

These examples show that the ^{238}Pu content of power reactor fuel can be dramatically increased with relatively minor changes to the basic nuclear fuel cycle. Plutonium-238 contents in the range of 8 to 10% are readily achieved, and values approaching 20% may be possible. The resulting plutonium isotopic mixture, and perhaps ^{237}Np, could be recycled in LWRs or utilized as a "safe" fuel for the core of a breeder, possibly in a non-weapons nation.

POTENTIAL HEATING PROBLEMS

Plutonium-238 decays by alpha emission with an 89-year half-life. The specific heat generation is very high, i.e., 0.56 W per gram of ^{238}Pu. Thus a 10-kg mass of plutonium (a sphere $\sim$4 in. diam) containing 10% ^{238}Pu would generate 560 W of heat. In air, a 150-W light bulb, of about the same size, is too hot to hold. Estimates have been made of the equilibrium interface temperature of such a 10-kg mass when insulated by varying thicknesses of a typical high explosive. The resulting temperatures are far above the critical temperature of most explosives.

The isotopic compositions of plutonium from three potential fueling scenarios are presented in Table 2. In case 1 the composition is representative of the first cycle discharge of standard PWRs, or a stowaway cycle. Case 2 represents the long-term buildup for complete recycle of uranium, plutonium, and neptunium (Scenario 1 of Fig. 1). Case 3 represents the long-term builup for the thorium-^{235}U cycle when uranium, plutonium, and neptunium are recycled (Scenario 2 of Fig. 1).

Table 2. Representative discharge plutonium isotopic concentrations

Isotope	Case 1[a]	Case 2[b]	Case 3[c]
^{236}Pu	1.39×10^{-7}	8.45×10^{-7}	1.29×10^{-6}
^{238}Pu	0.0147	0.1174	0.1854
^{239}Pu	0.5645	0.4964	0.3867
^{240}Pu	0.2647	0.2299	0.1877
^{241}Pu	0.1154	0.1251	0.1207
^{242}Pu	0.0407	0.1202	0.1095

[a]Case 1 is first-cycle discharge of standard PWR.
[b]Case 2 is fifth-generation discharge of Scenario 1 of Fig. 1 and Table 1.
[c]Case 3 is fifth-generation discharge of Scenario 2 of Fig. 1 and Table 1.

The heat generation rates of the plutonium compositions described in Table 2 are presented in Table 3. For comparison, the heat generation rates of the fission products and actinides in spent LWR fuel are also given. The specific heat output (per gram of plutonium or of total heavy metal, respectively) is about the same for plutonium produced in present LWRs (Case 1) and for typical spent reactor fuel after one year of cooling The "denatured" plutonium of Cases 2 and 3 generates 6 to 10 times as much heat, depending on the ^{238}Pu content.

A sufficiently ingenious organization with a highly developed technology could conceivably make a weapon out of plutonium which self-generates such amounts of heat. At the very least, though, the metal purification, shaping, and assembly steps of manufacture would become sufficiently difficult, and re-storage and delivery of the weapon so uncertain, that a fledgling weapons-maker might choose some other route. In principle, such problems may be circumvented by elaborate technology, but this technology is clearly beyond the capability of the unsophisticated weapons-maker, thereby providing an effective deterrent with respect to terrorists, criminal groups, and developing nations. Such technology, for the higher ^{238}Pu contents, may well be beyond the capability of all but experienced weapons design teams, thereby solving the deterrent problem in total since they already have weapons capability. In any case, other fissile materials would be more desirable.

NEUTRON AND GAMMA RADIATION

It is well known that the isotopes of plutonium are radioactive; all except ^{241}Pu (a beta emitter) decay by alpha emission. Additionally, all of them, especially the even-mass isotopes, have significant spontaneous fission rates. Further, in the presence of light elements (e.g., oxygen and fluorine), a significant (α,n) reaction occurs. The isotopes ^{236}Pu and ^{241}Pu also decay to daughters having high gamma activities. It is apparent that nearly all of the radiation characteristics are made more troublesome as the ^{238}Pu concentration is increased.

Table 4 presents the estimated gamma radiation dose rates at a distance of 1 ft, as a function of time after purification of a 10-kg mass of plutonium metal for each of the three cases described above.

Table 3. Decay heat generation rates

Material	Watts per gram[a] after cooling times of 1 year	2 years
Discharged fuel:		
Fission products	0.0106	0.00555
Actinides	0.00054	0.00033
Total	0.0111	0.0059
Purified plutonium:		
Case 1[b]	0.0116	0.0117
Case 2[c]	0.0685	0.0689
Case 3[d]	0.106	0.107

[a]Based on total heavy metal in material specified.
[b]Case 1 is first-cycle discharge of standard PWR.
[c]Case 2 is fifth-generation discharge of Scenario 1 of Fig. 1 and Table 1.
[d]Case 3 is fifth-generation discharge of Scenario 2 of Fig. 1 and Table 1.

Table 4. Gamma radiation dose rate (in mR/hr) at 1 ft from a 10-kg plutonium sphere

Time (days)	Case 1[a]	Case 2[b]	Case 3[c]
0.5	25	126	191
5	26	126	191
30	27	127	192
90	30	130	195
183	34	135	200
365	43	145	209
1095	79	183	245

[a]Case 1 is first-cycle discharge of a standard PWR.
[b]Case 2 is fifth-generation discharge of Scenario 1 of Fig. 1 and Table 1.
[c]Case 3 is fifth-generation discharge of Scenario 2 of Fig. 1 and Table 1.

Table 5. Neutron dose rate (mrem/hr) at 1 ft from 10-kg spheres

	Case 1[a]	Case 2[b]	Case 3[c]
Plutonium oxide	119	446	640
Plutonium metal	131	259	300

[a]Case 1 is first-cycle discharge of a standard PWR.
[b]Case 2 is fifth-generation discharge of Scenario 1 of Fig. 1 and Table 1.
[c]Case 3 is fifth-generation discharge of Scenario 2 of Fig. 1 and Table 1.

Table 5 presents the corresponding neutron dose rates also at a distance of 1 ft from 10-kg plutonium oxide or metal spheres. These rates are essentially independent of time, although small changes would be observed, primarily resulting from the (α,n) reaction from the buildup of ^{241}Am. A significant portion of this neutron dose results from neutron multiplication within the plutonium mass. The contribution of fission gammas has not been included in the estimates of gamma dose in Table 4. From Tables 4 and 5 it is apparent that most of the biological dose from handling kilogram quantities of plutonium will result from neutrons, but the rates are low enough that, by themselves, they do not provide an effective deterrent. However, the high neutron emission would cause weapon yields to be uncertain and strongly favors the realization of "fizzle" yield.

COMPLICATIONS TO THE POWER REACTOR FUEL CYCLE

The fuel cycle modifications suggested in this work are reasonably modest and involve little extension of projected technology. This is especially true if the thorium-denatured uranium cycle is considered as the base fuel cycle. These modifications are anticipated to increase the fuel cycle cost beyond that projected for a traditional plutonium recycle mode. Complications would be introduced in the following portions of the nuclear fuel cycle:

1. Shipment and handling of unirradiated mixed oxide fuel would become more difficult, but not nearly as difficult and expensive as for highly gamma spiked recycle fuel or spent fuel.
2. The greater amounts of ^{238}Pu would make it necessary to reexamine licensing and the environmental restrictions on all portions of the fuel cycle.
3. Nearly all portions of the LWR nuclear fuel cycle will be more expensive than the corresponding traditional plutonium recycle mode. These added costs would be especially apparent in enrichment, natural uranium, shipping, and fabrication. Smaller added costs would be incurred in reprocessing and waste handling. However, other suggested proliferation-resistant fuel cycles may also have significant impacts.

4. Fuel reprocessing would require the (partial) coprocessing of uranium and plutonium, primarily to keep specific heat and radiation levels in recovered products as low as possible and to utilize all of the ^{236}U in new fuel loadings. Neptunium, and possibly americium and curium, would be recovered either with the coprocessed uranium and plutonium or separately. The increased ^{238}Pu content would present some problems in reprocessing. The alpha radiation intensity of one-year-cooled fuel would be greater by up to a factor of 2, but the total radiation decay energy (including fission products) would be increased by less than 10%. Such radiation levels are expected to be manageable, especially with some degree of coprocessing which dilutes plutonium with uranium.
5. Fuel fabrication of plutonium-bearing fuels would require fully remotely operated facilities (which may be required in any recycle case). In refabrication of 20% plutonium--80% mixed oxide fuel, the specific alpha and heat generation rates would be about 1.5 ($\pm$ 0.4) times as great as from conventional LWR first-cycle plutonium. This is not expected to have a significant impact. In contrast, serious problems would be anticipated if the plutonium were concentrated into a reasonably pure and concentrated product, because the heat generation rate would be 5 to 10 times greater than for LWR first-cycle plutonium.

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