Reports

Radioactive Waste: The Problem of Plutonium

Abstract. *Systems under development for the recovery of plutonium from spent fuel and its recycling in fresh fuel would not significantly reduce either the total α-activity or the amount of fissile plutonium in radioactive waste relative to what is possible with once-through fuel cycles.*

In April 1977, as a result of increasing concerns that the recovery and recycling of plutonium from "spent" nuclear reactor fuel could ease its diversion to weapons uses, the U.S. government deferred plans for the commercialization of plutonium recycling technologies and urged other governments to do likewise until alternative, possibly more proliferation-resistant nuclear fuel cycles had been examined. Nevertheless, proposals to continue indefinitely the present reliance on once-through fuel cycles have drawn a number of criticisms:

1) Without the savings in uranium that plutonium recycling would make possible, the world's resources of high-grade uranium ore will be more rapidly exhausted.

2) The long-term toxicity of radioactive wastes will be significantly increased if the plutonium is not recycled and "burned up."

3) If the spent fuel is buried, the repository sites will become potential "plutonium mines" for future groups desiring nuclear weapons material.

Issue 1 has already been dealt with elsewhere (*1*). In this report we discuss issues 2 and 3 and show that the quantities of plutonium and other long-lived α-emitters in the spent fuel from a once-through fuel cycle need be no larger than those projected for the radioactive wastes from the standard plutonium recycling systems.

The radiotoxicity and thermal energy output of spent light-water reactor (LWR) fuel after a few hundred years will be dominated by the α-decays of a few heavy transuranic radionuclides (*2*, *3*). The toxicity of the transuranics is very uncertain, however, because, depending upon their chemical state, the wall of the human gastrointestinal tract may or may not be a very effective barrier to their absorption into the body (*4*). Even slight differences in water chemistry have been found to change the potential uptake of plutonium by orders of magnitude, for example (*5*). We therefore do not follow here the usual procedure of measuring the toxicity of the transuranics by using official ingestion limits. Instead we have chosen to quantify the presence of the transuranics and other α-emitters by a physical measure, the total level of α-activity (*6*).

Figure 1a shows the total α-activity in the waste streams from the generation of 1 gigawatt-year of electric energy [1 GW-year (e)] by the current low-enriched uranium once-through LWR fuel cycle compared with the corresponding α-activities in the wastes from three alternative fuel cycles. Two of these alternatives involve the recycling of plutonium in LWR's and liquid-metal fast breeder reactors (LMFBR's). These are the fuel cycles on which the most development work has been done during the past decade. We have assumed that a total of 2 percent of the plutonium and uranium and 100 percent of all other α-emitters present in the spent fuel prior to reprocessing find their way into the fuel reprocessing and refabrication wastes (*7*). The third alternative fuel cycle is a once-through high-temperature, gas-cooled reactor (HTGR) system, which has been designed to be highly uranium-efficient and to produce a minimum of plutonium subject to the constraint that the fuel does not contain other chemically separable weapons-usable material (see below).

It will be seen from Fig. 1a that plutonium recycling in LWR's and LMFBR's reduces the α-activity in the wastes by factors of only about 2 and 4, respectively, relative to the standard once-through LWR fuel cycle for the period during which the toxicity of the wastes is dominated by the transuranics (between approximately a few hundred and 10^5 years after discharge from the reactor). This is

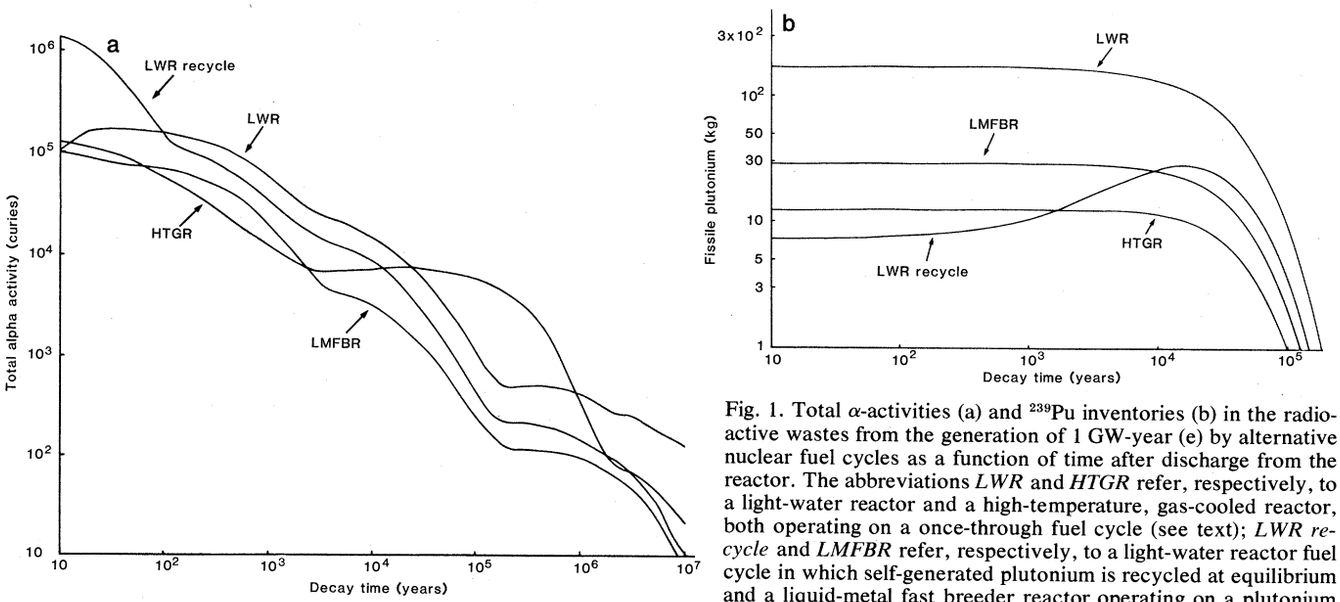

Fig. 1. Total α-activities (a) and ^{239}Pu inventories (b) in the radioactive wastes from the generation of 1 GW-year (e) by alternative nuclear fuel cycles as a function of time after discharge from the reactor. The abbreviations *LWR* and *HTGR* refer, respectively, to a light-water reactor and a high-temperature, gas-cooled reactor, both operating on a once-through fuel cycle (see text); *LWR recycle* and *LMFBR* refer, respectively, to a light-water reactor fuel cycle in which self-generated plutonium is recycled at equilibrium and a liquid-metal fast breeder reactor operating on a plutonium fuel cycle.

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comparable to or less than the reduction that is possible for decay times shorter than 10^4 years if one were to use the alternative, once-through fuel cycle.

The surprisingly small reduction of α-activities realized by recycling is due to the fact that plutonium is exposed in recycling to more neutron bombardment than in once-through fuel cycles. As a result, a significant fraction of the long-lived α-activity in the wastes from recycling is associated with transuranics (americium in particular) built up by neutron capture on plutonium. This effect is considerably reduced in the LMFBR plutonium fuel cycle because of the significantly reduced probability of nonfission capture by the transuranics of "fast" (energetic) LMFBR neutrons. But, since the LMFBR fuel cycle would recycle about twice as much plutonium per gigawatt-year of electric energy as the LWR, twice as much plutonium would be lost to waste (for the same fractional losses).

The initial α-activity of the once-through thorium-uranium HTGR fuel is lower than that of the once-through uranium fuel. After 10^4 years, however, the α-activity of the thorium-based fuel declines more slowly because the product of neutron capture on thorium, ^{233}U (half-life, 1.62×10^5 years), has a longer half-life than ^{239}Pu (half-life, 0.24×10^5 years).

The concern about spent fuel repositories becoming "plutonium mines" stems from the fact that a 1000-MW (e) LWR on the current once-through fuel cycle discharges about 175 kg of ^{239}Pu per full power year [that is, for each gigawatt-year of electric energy produced]. Only about 10 kg would be required to make the equivalent of the bomb that destroyed Nagasaki (*8*). It is therefore of interest to consider the extent to which plutonium recycling would lessen this problem.

In Fig. 1b we compare the ^{239}Pu in the waste from 1 GW-year (e) of fission power by a once-through LWR fuel cycle with the corresponding ^{239}Pu inventories in the wastes from plutonium recycling in LWR's and LMFBR's and from the once-through HTGR fuel cycle with low ^{239}Pu production. Plutonium recycling in both LWR's and LMFBR's reduces the ^{239}Pu in the wastes by approximately one order of magnitude relative to the once-through LWR case. This is less than might be naïvely expected on the basis of the 2 percent plutonium loss rate to waste assumed for the plutonium recycling cases. The inventories of ^{239}Pu in the spent fuel are two (LWR) and eight (LMFBR) times larger for the recycling systems than for the once-through LWR cycle, however, and, especially in the case of the LWR recycle wastes, there are substantial inventories of ^{243}Am (half-life, 7300 years), which decays into ^{239}Pu (hence the rise in the amount of ^{239}Pu in the wastes from this fuel cycle after 1000 years).

The curve for the once-through HTGR fuel cycle in Fig. 1b indicates that there are alternatives to plutonium recycling that accomplish a similar reduction in the quantity of ^{239}Pu in radioactive wastes. In this case the reduction has been achieved principally by reducing the concentration of ^{238}U in the fuel (^{239}Pu is created by neutron capture on ^{238}U). This approach has been pursued only to the point where the ^{235}U enrichment of the uranium in the fresh LWR fuel rises to 20 percent, because highly enriched uranium is itself nuclear weapons–usable material. For an increase from the 3 per cent enrichment characteristic of the LWR once-through fuel cycle, however, this corresponds to a sevenfold reduction in the amount of ^{238}U associated with a given amount of ^{235}U in the fuel. The fuel-extending value of the displaced ^{238}U is maintained by substituting thorium (*9*). An additional, although less dramatic, reduction in the ^{239}Pu content of the spent HTGR fuel has been achieved because the HTGR fuel is driven to high "burnups" (measured in terms of the thermal energy release per kilogram of heavy metal in the fuel). This results in an increased fraction of the ^{239}Pu produced in the fuel being fissioned in place or being converted into higher transuranics.

Thus reductions in the plutonium and overall transuranic content of radioactive wastes comparable to those that are achieved in the standard plutonium recycling systems can be accomplished with refined once-through fuel cycles. These findings apply to the plutonium recycling technology that has been developed to date. Future technology might reduce the quantities of plutonium and other transuranic elements in the radioactive waste streams from recycle systems to levels well below those achievable with once-through systems. The plutonium in recycle systems passes through so much plumbing and machinery, however, and goes through so many changes in chemical and physical state that reducing losses by a large amount may well be quite costly. Furthermore, the transmutation of the average higher transuranic atom into shorter-lived fission products in reactors would take many cycles. Even with no leakage, it would take 100 years of very careful recycling in a breeder reactor to destroy 90 percent of the neptunium, americium, and curium that would otherwise go into the reprocessing wastes (*10*).

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References and Notes

1. H. A. Feiveson, F. von Hippel, R. H. Williams, *Science* **203**, 330 (1979).
2. For a recent review, see "Report to the American Physical Society [APS] by the study group on nuclear fuel cycles and waste management," *Rev. Mod. Phys.* **50**, S110 (1978). This study also overlaps this report in that it compares the ingestion toxicities of (i) spent low-enriched uranium fuel from an LWR, (ii) high-level waste from an LWR operated with self-generated plutonium recycling, and (iii) spent fuel from a HTGR fueled with a mixture of high-enriched uranium and thorium. The APS study finds a range of toxicities for these wastes differing by approximately 30 after 1000 years and concludes (p. S112) that differences of these magnitudes do "not give a strong incentive for choosing one alternative fuel cycle over another."
3. For a detailed analysis, see H. Krugmann, thesis, Princeton University (1978).
4. A. Lindenbaum, J. Lafuma, M. W. Rosenthal, W. S. Snyder, D. M. Taylor, R. C. Thompson, M. Izawa, Yu I. Muskalev, J. M. Vaughan, *The Metabolism of Compounds of Plutonium and Other Actinides* (International Commission on Radiological Protection Publication 19, Pergamon, New York, 1972).
5. See, for example, R. P. Larsen and R. D. Oldham, *Science* **201**, 1008 (1978); see also M. F. Sullivan, J. L. Rayn, L. S. Gorham, K. M. McFadden, in *Pacific Northwest Laboratory Annual Report for 1978 to the Department of Energy Assistant Secretary for Environment*, Part 1, *Biochemical Sciences* (Pacific Northwest Laboratory, Seattle, 1979), p. 392.
6. Shifting from ingestion toxicity to α-activity does not change significantly the relative weights of the various transuranic α-emitters, since they are ordinarily assumed to have approximately the same ingestion toxicity per curie [*Basic Safety Standards for Radiation Protection* (Safety Series No.9, International Atomic Energy Agency, Vienna, 1967)]. It does lead, however, to an increased weighting of the transuranics relative to some natural α-emitters (^{226}Ra in particular) which, for the chemical states assumed in obtaining the standard toxicity values, are much more readily absorbed into the body from the gastrointestinal tract than the transuranics. Since the α-emissions dominate the thermal energy output of the waste after the first few hundred years and each α-particle carries approximately the same amount of energy (between 4×10^6 and 7×10^6 eV), the level of α-activity is also a good measure of the long-term heat generation by the waste.
7. The isotopic inventories in the LWR (which we assume for specificity is a pressurized water reactor) and LMFBR spent fuel are taken from C. W. Kee, A. G. Croft, and J. O. Blomeke [*Updated Projections of Radioactive Wastes to Be Generated by the U.S. Nuclear Power Industry* (Publication ORNL/TM 5427, Oak Ridge National Laboratory, Oak Ridge, Tenn., 1976)]. The spent fuel isotopic inventory for the once-through HTGR fuel cycle is taken from E. Teuchert [*"Once Through" Cycles in the Pebble Bed HTR* (Publication Jül-1470, Kernforschungsanlage Jülich, Jülich, West Germany, 1977)] for an initial $^{232}Th/^{238}U$ ratio of 1.9, a uranium enrichment of 20 percent, and an average fuel "burnup" of 100 MW-day (thermal) per kilogram of heavy metal. Since Teuchert does not give the inventories of all the α-emitters of interest, the missing ones were derived from those given by H. J. Rütten [*Daten abgebrannter Brennelemente verschiedener Reaktoren und Brennstoffzyklen ermittelt mit dem Computercode ORIGEN-EXJÜL* (internal report KFA-IRE-IB-12/79, Kernforschungsanlage Jülich, Jülich, West Germany, 1979)]. Rütten's numbers are for 36 percent higher burnup fuel than

Teuchert's. Each of his inventories of α-emitters created by neutron capture was therefore reduced by the ratio of the inventories (Teuchert's/Rütten's) of the nearest precursor listed by both authors on the dominant neutron build-up chain leading to the isotope in question. This procedure leads to a slight overestimate of the HTGR spent fuel α-activity and ^{239}Pu inventory.

8. We ignore the fissile isotope ^{241}Pu here since it has such a short half-life (13 years). The critical spherical mass of ^{239}Pu diluted with up to an equal amount of nonfissile ^{240}Pu + ^{242}Pu in a metallic α-phase surrounded by a thick uranium neutron reflector is 4 to 5 kg. The complete fissioning of 1 kg of heavy metal would yield energy approximately equal to the explosion of 20,000 tons of high explosive [T. B. Taylor, *Ann. Rev. Nucl. Sci.* **25**, 407 (1975)].
9. Some of the ^{238}U is transmuted by neutron capture and subsequent radioactive transformations into ^{239}Pu, some of which is fissioned in place. Thorium-232 is similarly converted into fissile ^{233}U. Pure ^{233}U is weapons-usable but differs from ^{239}Pu in that, like ^{235}U, it will be "isotopically denatured" for nuclear-weapons purposes as a result of its dilution by the ^{238}U in the fuel [see (*1*)].
10. See figure 7D1 in (*2*), p. S116.

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Carbon Budget of the Southeastern U.S. Biota: Analysis of Historical Change in Trend from Source to Sink

Abstract. *Documentation of settlement patterns and deforestation in the southeastern United States allows evaluation of regional carbon dynamics since A.D. 1750. From 1750 to 1950, the Southeast was a net source for carbon at an average rate of 0.13 gigaton per year. Only in the past 20 to 30 years has increased productivity of commercial forests resulted in a sink for atmospheric carbon dioxide of 0.07 gigaton per year.*

The observed increase in atmospheric CO_2 over preindustrial levels has been ascribed largely to burning of fossil fuels (*1*) and to forest clearing and burning (*2*). We examined the carbon dynamics of the southeastern United States over the past two centuries in order to estimate the presettlement carbon pool, document carbon losses from deforestation, and determine whether this region is a source or a sink of atmospheric CO_2. Such reconstructions provide insight into the source or sink strength of the terrestrial biosphere through time and suggest a future trajectory of carbon dynamics under given conditions of land use (*3*).

U.S. agricultural census statistics (*4*) and forest survey records (*5*) provide data for determining changes in land use during the settlement of Alabama, Arkansas, Delaware, Florida, Georgia, Kentucky, Louisiana, Maryland, Mississippi, North Carolina, South Carolina, Tennessee, Virginia, and West Virginia, an area of 1,405,737 km^2 (*5*), or 12 to 16 percent of the world's temperate forest area (*2*). Before settlement (A.D. 1750) (Fig. 1), 91.6 percent of the land area of these states was forest, 3.8 percent was prairie, and 4.6 percent was marsh (*6*). Forests and native prairies were rapidly converted to agricultural land as settlers pushed the frontier westward during the late 1700's and early 1800's (*4*, *7*). By 1880, less than 35 percent of the Southeast remained in virgin forest—principally in the southern Appalachian Mountains, Mississippi River bottomlands, Ozark and Ouachita mountains of Arkansas, and Gulf Coastal Plain pinelands (*8*). The coastal plain and southern Appalachian forests were logged between 1880 and 1920 (*9*). Southeastern bottomland forests are expected to be converted largely to cropland before 1990 (*10*).

However, since 1950, clearing of the bottomlands has been offset by upland old-field succession, particularly in the Piedmont (*11*). Intensively managed commercial (secondary) forest holdings have increased in size as farm woodlots have diminished (Fig. 1). Today, virgin forests occupy less than 1 percent of their former area (Fig. 1), persisting only in small isolated stands.

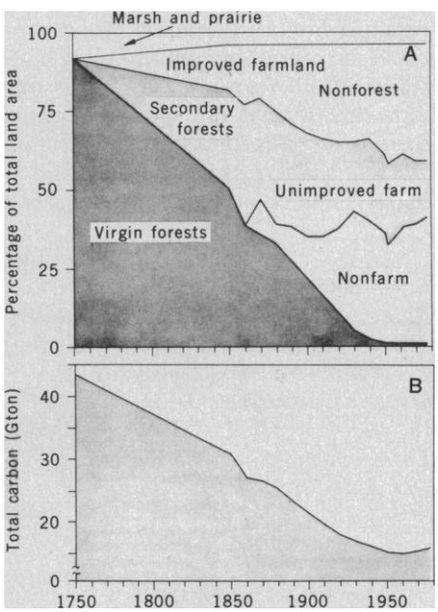

Fig. 1. (A) Land-use history and (B) changes in total carbon for the southeastern United States, A.D. 1750 to 1977. The areas in forest and rainforest for 1950 are based on the natural vegetation map of Shantz and Zon (*6*). The data for 1850 to 1950 are from (*4*), with improved farmland defined as cleared land in cultivation or pasture and unimproved farmland defined as including pastured and unpastured woodlots. The data for 1952 to 1977 are based on inventories by the U.S. Forest Service (*5*), with the nonfarm secondary forest category including commercial forests in national and state forests, paper and lumber company holdings, and other private ownerships (excluding farmland).

Estimates of timber volume for virgin forests of 1880 (*8*) and for secondary forests of 1952, 1962, 1970, and 1977 (*5*) were used to estimate total live (aboveground and belowground) biomass and total carbon (including detrital soil carbon) on forested land (*12*). The aboveground biomass of the virgin forests averaged 343 Mg/ha, total live biomass was estimated as 460 Mg/ha, and total carbon averaged 327 Mg/ha (*12*). The aboveground biomass figure, estimated from Sargent (*8*), is less than that estimated by Whittaker (*13*) for undisturbed cove hardwood forests (500 to 600 Mg/ha), but greater than that for old-growth commercial forests of the southern Appalachians (176 Mg/ha) (*14*). The cove forests examined by Whittaker represent an upper limit, a potential not attained uniformly across the presettlement landscape because of various disturbances (*15*). Biomass on agricultural land was calculated by using the production values of DeSelm (*16*). Land clearing and cultivation were estimated to diminish soil carbon by 40 percent (*16*, *17*).

From 1750 to 1960, total carbon in soil and vegetation decreased nearly linearly from 43.3 to 15.1 Gton (Fig. 1 and Table 1). Replacement of nearly 55 percent of the original forest land with secondary forests did not restore carbon reserves depleted by extensive agricultural utilization over that 210-year period. With an average release of 0.13 Gton of carbon per year (Table 1), the southeastern United States has served as a major carbon source to the atmosphere during most of the time since the Industrial Revolution.

Between 1952 and 1977, the aboveground biomass on commercial forest land increased from 53.2 to 72.2 Mg/ha (*5*). This gain reflects the increase in holdings of intensively managed forest land (Fig. 1), on which net annual growth has increased due to reforestation of nonstocked areas and control of species composition and stand density (*10*). Increased storage of wood in planted or early successional commercial forest stands has resulted in a net increase of yield over harvest in the past 20 to 30 years (*18*). This, in the context of overall stabilization of forest and nonforest land areas, has reversed the trend in carbon

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