Science and Global Security, 13:169–181, 2005

ISSN: 0892-9882 print
DOI: 10.1080/08929880500357682

Taylor & Francis
Taylor & Francis Group

Limited Proliferation-Resistance Benefits from Recycling Unseparated Transuranics and Lanthanides from Light-Water Reactor Spent Fuel

Jungmin Kang[1] and Frank von Hippel[2]

[1]Seoul, Republic of Korea
[2]Program on Science and Global Security, Princeton University, Princeton, NJ

Keeping LWR plutonium mixed with other transuranics and with lanthanide fission products other than ^{154}Eu does not make it significantly more self protecting or more difficult to fabricate into a nuclear weapon. Gamma-ray and neutron doses at one meter, heat generation, and spontaneous-neutron emission are calculated from 1-kg metal balls of weapon-grade plutonium, reactor-grade plutonium, and the full mix of transuranics in high-burnup light-water-reactor (LWR) spent fuel with and without the lanthanide fission products from the spent fuel. The total radiation dose rate from transuranics without the lanthanides is more than three orders of magnitude lower than the IAEA's threshold for self-protection, 1 Sv/hr (100 rems/hr) at 1 meter. Inclusion of either of two lanthanide fission products, ^{144}Ce and ^{154}Eu, could increase the dose rate above the self-protection threshold. However, ^{144}Ce has a half-life of only 0.8 years and has already decayed away in all but the most recently discharged LWR spent fuel. ^{154}Eu has a half-life of nine years but is not recycled with the transuranics in the pyroprocessing fuel cycle that was developed for the Integrated Fast Reactor.

Received 3 January 2005; accepted 10 April 2005.
We are grateful for very helpful reviews of two drafts by Steve Fetter. We would also like to thank Edwin S. Lyman, who first pointed out to us that ^{154}Eu does not follow the transuranics in pyroprocessing.
Address correspondence to Frank N. von Hippel, Program on Science and Global Security, 221 Nassau St, 2nd Floor, Princeton University, Princeton, NJ 08542-4601. E-mail: fvhippel@princeton.edu and Jungmin Kang, 212-203 Shinbanpo 4 Cha Apt., Jamwon-dong, Seocho-gu, Seoul 137-949, Republic of Korea. E-mail: jmkang55@hotmail.com

BACKGROUND

After India's 1974 nuclear test, which used plutonium separated using declassified technology and training and equipment provided by the United States,[1] the Ford and Carter Administrations decided to oppose further export of reprocessing technology to countries of proliferation concern. They also decided that plutonium recycle was not required in the near term by civilian nuclear-power programs and that the United States could therefore store spent power-reactor fuel.

In 1993, the Clinton Administration restated this antireprocessing policy as follows, "The United States does not encourage the civil use of plutonium and, accordingly, does not itself engage in plutonium reprocessing."[2]

The George W. Bush Administration has relaxed this antireprocessing policy somewhat. According to its 2002 National Security Presidential Directive on National Strategy to Combat Weapons of Mass Destruction:

> the United States will continue to discourage the worldwide accumulation of separated plutonium and to minimize the use of highly-enriched uranium. As outlined in the National Energy Policy, the United States will work in collaboration with international partners to develop recycle and fuel treatment technologies that are cleaner, more efficient, less waste-intensive, and more proliferation resistant.[3]

Accordingly, in 2003, the U.S. Department of Energy's Office of Nuclear Energy (DOE-NE) launched the Advanced Fuel Cycle Initiative (AFCI), which then DOE Secretary Spencer Abraham described as, "research that can optimize the use of the first repository [Yucca Mountain] and possibly reduce the need for future repositories."[4]

The concern about U.S. repository capacity, as elaborated in the DOE-NE's 2003 report to Congress on the AFCI, is that

> the statutory limit for the planned [Yucca Mt] geological repository, 63,000 Mt [metric tons] of civilian nuclear spent fuel, will be reached in 2015. . . . As a result, the quantity of spent fuel produced by nuclear power plants may become a long-term challenge to the possibility of building new nuclear power plants as anticipated by the *National Energy Policy.*[5]

DOE-NE is therefore developing a strategy to reprocess U.S. spent fuel that would first extract pure uranium from dissolved spent fuel and then process, "mixtures of plutonium and selected minor actinides for preparing proliferation-resistant fuels. . . . If implemented successfully, this treatment technology could significantly reduce the cost of the first repository and potentially eliminate the technical requirement for a second."[6]

The accumulation of decay heat in the rock between the emplacement tunnels, in the period after which forced ventilation ceases, limits the capacity of an above-ground-water-level repository. With the removal of the long-lived transuranics, the cumulative output of thermal energy by the radioactive waste in the first few thousand years would be significantly reduced. For Yucca Mountain, it has been estimated that removing the transuranics would make it possible to store the remaining radioactive waste from about five times as much spent fuel as could be stored there directly without reprocessing.[7]

The DOE has been sensitive—rhetorically, at least—to the problem that drove the U.S. away from reprocessing in the first place and insists that, "UREX . . . can reduce the proliferation risk by avoiding the separation of plutonium from other radioactive species—thereby rendering the plutonium unusable for weapons applications."[8]

The current version of UREX ("UREX+") that is being developed, however, would keep the plutonium product mixed only with the relatively small quantity of neptunium-237 present in spent fuel.[9] ^{237}Np does not make the mix more self-protecting because it is more weakly radioactive than plutonium and has approximately the same critical mass as weapon-grade uranium.[10]

Another "proliferation-resistant" reprocessing technology that has attracted favorable attention from the Bush Administration is "pyroprocessing," an electrochemical process that was developed in connection with Argonne National Laboratory's proposal for an Integral Fast Reactor. Each group of IFRs would have its own integrated small reprocessing and fuel refabrication facility. Vice President Cheney's 2001 Energy Task Force endorsed this reprocessing technology in its *National Energy Policy* report:

> The United States should reexamine its policies to allow for research, development, and deployment of fuel conditioning methods (such as pyroprocessing) that reduce waste streams and enhance proliferation resistance.[11]

In this article, we evaluate the proliferation resistance that would be added if plutonium were not separated from the other transuranic isotopes in spent LWR fuel, and then, if, as in pyroprocessing, the transuranics were not separated from the non-europium lanthanides.[12]

CHARACTERISTICS OF TRANSURANICS

Table 1 shows the composition of the transuranic mix in low-enriched uranium (LEU) fuel 20 years after the fuel has been discharged with a "burnup" (fission energy release) of 53 megawatt-days per kilogram of heavy metal (MWd/kgHM). It also shows the decay-heat production, spontaneous fission neutron emission rate, and gamma and neutron dose rates from a 1-kg metal

Table 1: Heat, neutron emission and doses from 1 kg of various pure transuranic isotopes, the mixture in 53 MWd/kg LEU spent fuel and weapon-grade plutonium (20 years after discharge).

Isotope	Halflife (years)	Bare critical mass (kg)*	Mass fract. in Trans U	Heat (W/kg)	Spont. Fiss. neutrons ($\times 10^5$/kg-sec)	Dose rate (10^{-5} Sv/hr-kg) at 1 m	
						Gamma	Neutron**
^{237}Np	2.14×10^7	57.***	0.066	0.022	0	0.08	—
^{239}Np (from ^{243}Am)	2.4 days	—	1.6×10^{-8}	5.6×10^5	0	3.9×10^8	
Pu-tot	—	14.4	0.824	19.9	4.6	0.52	0.33
^{241}Am	432.	60.0	0.089	114	0.012	126.	9×10^{-4}
^{242m}Am	141.	9.1	9.3×10^{-5}	3.84	1.49	33.	0.11
^{243}Am	7400.	208.8	0.018	6.4	0.03	38.	0.002
^{243}Cm	28.5	8.6	5.1×10^{-5}	1900.	—	60,000.	0.
^{244}Cm	18.1	27.0	3.7×10^{-3}	2830.	1.11×10^5	208.	7950.
^{245}Cm	8500	9.2	3.9×10^{-4}	5.7	-	81.	0.
TransU	—	17.9	1.000	37.3	414.	22.	30.
WgPu****	—	10.7		2.3	0.5	0.08	0.04
			Mass fract. in Pu				
^{236}Pu	2.9	6–8	2.3×10^{-9}	18,500.	349.	609.	25.
^{238}Pu	87.7	9.6–9.8	0.029	568.	26.6	13.8	1.9
^{239}Pu	2.4×10^4	10.1	0.555	1.92	2.3×10^{-4}	0.08	1.6×10^{-5}
^{240}Pu	6.5×10^3	36.9	0.266	7.1	9.1	0.19	0.65
^{241}Pu	14.4	13.0	0.064	3.2	—	0.18	0.
^{242}Pu	3.8×10^5	83.4	0.085	0.113	16.9	0.03	1.2

*For individual isotopes, from "Critical masses of bare metal spheres using SCALE/XSDRN" by R. Q. Wright, W. C. Jordan, and R. M. Westfall, Oak Ridge National Laboratory, *Proceedings of the Annual Meeting of the American Nuclear Society,* San Diego, June 4–8, 2000, p. 167. We have checked these numbers using MCNP and the cross-section library ENDF/B-VI.5 and find consistent results, except for ^{243}Am, where we find a critical mass of 144 kg. For Pu-tot, WgPu, and TransU, authors' calculations assuming mass density of 19.86 g/cc for Pu and 19 g/cc for TransU.

**Using a fluence-to-dose factor of 0.21×10^{-9} Sv-cm^2 for 2-MeV neutrons, *Neutron and gamma-ray fluence-to-dose factors*, Op. cit.

***"Neptunium nuclear data & criticality" by Mark Chadwick, presentation at the 53rd Cross section evaluation working group meeting & U.S. nuclear data program meeting, common session on nuclear data for homeland security, Brookhaven National Laboratory, Nov. 4–7, 2003, www.nndc.bnl.gov/nndc/proceedings/ 2003csewgusndp/homeland/04Chadwick.pdf

****^{238}Pu, 0.012%; 239Pu, 93.8%; ^{240}Pu, 5.8%; ^{241}Pu, 0.35%; and ^{242}Pu, 0.022%, "Explosive properties of reactor-grade plutonium" by J. Carson Mark, *Science & Global Security 4*: 111.

ball of each transuranic isotopic, weapon-grade plutonium, and the plutonium and transuranic mixes in the spent fuel.[13]

The composition of the fuel was calculated with ORIGEN 2.1,[14] assuming an initial LEU fuel enrichment of 4.4 percent ^{235}U. The gamma radiation doses were calculated using the American Nuclear Society's gamma-ray fluence-to-dose factors.[15] The self-shielding of the metal ball was calculated for a density of 19.86 g/cc using the Los Alamos MCNP4C2 Monte Carlo particle-transport code.[16] Some of the significant gamma-dose contributions were checked by hand.[17]

The combined gamma and neutron dose rate from the 1-kg transuranic sphere is about 0.5 mSv/hr—three orders of magnitude lower than the 1 Sv/hr

level that, according to the IAEA, justifies a reduced level of physical protection.[18] Although these radiation levels would significantly increase cancer risks among workers exposed to them year round, it must be assumed that, in a national or terrorist weapons program, the plutonium could be separated chemically from the transuranic mix without radiation shielding.

Thus the admixture of nonplutonium transuranics does not increase the proliferation-resistance of the plutonium significantly more than would a slight dilution with chemically similar nonradioactive material.

It should be noted also that ^{241}Am and ^{243}Am, which, with ^{237}Np, comprise the bulk of the nonplutonium material in the transuranic mix, are considered alternative nuclear materials and have bare critical masses comparable to some of the isotopes that are present in similar concentrations in reactor-grade plutonium (see Table 1).

Classified studies have concluded that, if a group could construct a nuclear weapon with weapon-grade plutonium, it could also construct one from civilian "reactor-grade" plutonium such as the mix of plutonium isotopes shown in Table 1:

> [A] subnational group using designs and technologies no more sophisticated than those used in first-generation nuclear weapons could build a nuclear weapon from reactor-grade plutonium that would have an assured, reliable yield of one or a few kilotons (and a probable yield significantly higher than that).[19]

Because much of the nuclear-energy establishment still does not accept this conclusion, we briefly review some of the elements of the analysis behind it.

Preinitiation. As shown in Table 1, the neutron-emission rate from reactor-grade plutonium is about an order of magnitude greater than that from weapon-grade plutonium, and the neutron-emission rate of the transuranic mix is about two orders of magnitude higher than from reactor-grade plutonium alone (because of the high spontaneous fission rate of ^{244}Cm). The high rate of neutron emission from reactor-grade plutonium and the transuranic mix have led some observers to conclude that these materials are unusable in nuclear weapons.

The probable explosive yield from a Nagasaki-type solid-core implosion weapon does depend on the rate of emission of spontaneous fission neutrons. The implosion from subcriticality to maximum supercriticality takes about 10 microseconds. If a chain reaction begins before maximum compression (preinitiation), the expected yield will be reduced from about 20 kilotons to as low as one kiloton. This is the reason the ^{240}Pu content of the plutonium used in the Nagasaki bomb was minimized. However, no level of spontaneous neutron emission would reduce the yield below about 1 kiloton.[20] A one-kiloton explosive would still be a devastating weapon.

Classified studies have also concluded that the designs of modern nuclear weapons are insensitive to preinitiation:

> [A]dvanced nuclear weapon states such as the United States and Russia, using modern designs, could produce weapons from reactor-grade plutonium having reliable explosive yields, weight, and other characteristics generally comparable to those of weapons made from weapon-grade plutonium.[21]

From a neutronics point of view, therefore, the transuranics mix would not be significantly inferior to reactor-grade plutonium as a weapons material.

Heat management. Table 1 also shows that the rate of heat release in reactor-grade plutonium is almost an order of magnitude greater than from weapon-grade plutonium. For eight kilograms of reactor-grade plutonium, an amount for which, according to weapon experts that advise the IAEA, "the possibility of manufacturing a nuclear explosive device cannot be excluded,"[22] the heat-generation rate would be about 160 watts. If this much plutonium were shaped into a solid sphere with a density of 16 g/cc, it would have a radius of only 6 cm and would become quite hot in air because of its small heat-transfer area. We estimate a surface temperature of 190°C for a bare sphere—much higher than the 60°C that would be reached by a similar sphere of weapon-grade plutonium.[23] Inside an insulating layer of explosives, the plutonium would heat up to much higher temperatures. Since most explosives become unstable above about 200°C,[24] this means that a nuclear warhead containing a small solid "pit" of reactor-grade plutonium would either have to have a cooling system or have a precooled pit inserted some hours before detonation, with the length of time depending upon the heat capacity of the tamper and other design details.[25] The Nagasaki bomb and other early nuclear weapons had an "insertable" pit design for safety and security reasons.[26]

The rate of heat emission from the transuranic mix is only about twice that of reactor-grade plutonium. Technical approaches that would allow the use of reactor-grade plutonium, such as cooling and insertable pits, therefore should work almost as well with the transuranic mix. Thus, from a heat-management point of view, the transuranic mix also should be considered as a potential direct-use nuclear-weapon material.

Critical mass. It will be seen from Table 1 that the fast-critical mass of reactor-grade plutonium is 1.35 times greater than that of weapon-grade plutonium and that of the transuranic mix is 1.7 times greater. These differences are smaller than the difference between the critical mass of weapon-grade plutonium and weapon-grade (94% enriched) uranium, which has a bare critical mass of about 50 kg.[27]

RADIATION DOSE RATES FROM LANTHANIDE FISSION PRODUCTS

The radioactive lanthanide fission products that contribute significantly to the activity of spent LWR fuel after a period of cooling of a few months or more are listed in Table 2.[28] Also shown are:

Table 2: Important lanthanide-series fission products in LEU spent fuel at discharge (53 MWd/kgHM).

Isotope	Halflife	Heat (Watts/gm)	Grams per kg transuranics	γs/decay (av. E_γMev)*	Gamma Dose rate (Sv/hr-kgHM at 1 m)
			In pyroprocessing product		
^{144}Ce	0.78 y.	2.12	32.2	0.13 (0.12)	0.10
^{144}Pr (from ^{144}Ce)	17.3 min.	5.6×10^{5}	1.37×10^{-3}	0.023 (1.2)	8.1
^{147}Pm	2.6 y.	0.33	10.9	3×10^{-5} (0.12)	10^{-4}
^{151}Sm	90 y.	0.003	1.54	3×10^{-4} (0.022)	2×10^{-8}
			Not in pyroprocessing product		
^{154}Eu	8.8 y.	2.42	6.16	1.5 (0.74)	2.31
^{155}Eu	4.7 y.	0.34	2.28	0.53 (0.094)	0.003

* *NuDat 2: Decay radiation database version of 8/12/2004, op. cit.*

- The grams of each isotope per kilogram of transuranics at the time of spent-fuel discharge from the reactor,
- The most important gamma emissions that accompany the decay of each lanthanide, and
- The contribution of each lanthanide to the radiation dose from a 1-kg sphere of transuranic metal if the sphere were made immediately after spent fuel discharge.

We have checked the dose rates from ^{144}Ce and ^{144}Pr by hand.[29] Only the doses from ^{144}Pm, the short-lived decay product of ^{144}Ce, and from ^{154}Eu are significant on the scale set by the IAEA's 1 Sv/hr at 1 m self-protection threshold.

Advocates of pyroprocessing have pointed to the proliferation-resistance benefits of not separating out the lanthanides from the transuranics. Pyroprocessing *does* separate out europium, however,[30] so the full burden of maintaining a high radiation field falls on ^{144}Pr and its parent radioisotope, ^{144}Ce.

Unfortunately ^{144}Ce has a relatively short halflife. As shown in Figure 1, even if all the ^{144}Ce follows the transuranics,[31] the radiation level from the ^{144}Ce in 1 kg of transuranic metal will have declined below the self-protection threshold a little more than two years after spent-fuel discharge. Within five years, it will be below 0.1 Sv/hr.

Therefore, the short-lived radiation barrier from ^{144}Ce would be relevant only for a fuel cycle involving separation and recycle of the transuranics in spent fuel *very* soon after spent-fuel discharge. IFR advocates believe this would be possible for a colocated IFR and pyroprocessing plant. A paper from Argonne National Lab on the proliferation-resistance of the IFR fuel cycle assumes recycle back into the reactor 100 days after fuel has been discharged from the reactor.[32] Using that assumption, it calculates a radiation field of

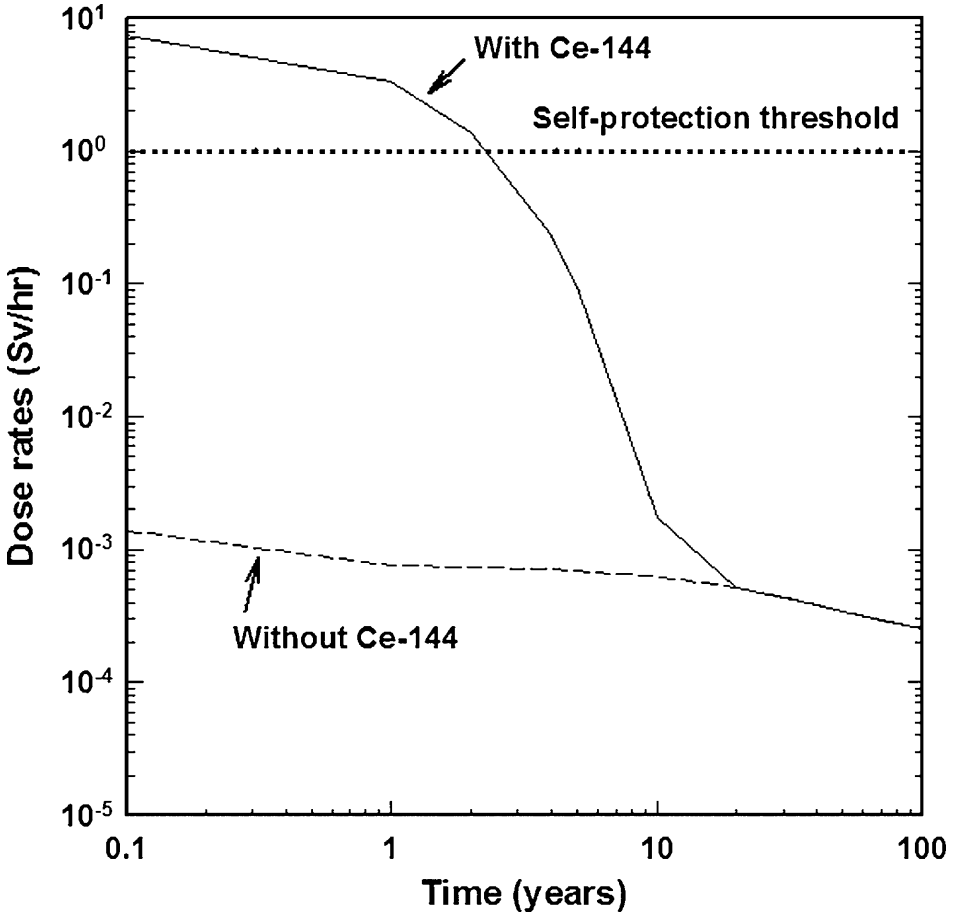

Figure 1: Effect of including ^{144}Ce on gamma-radiation dose rate 1 m from a 1-kg sphere of transuranics extracted from 53 Mwd/kgHM spent LEU fuel.

4 Sv/hr 1 m from 10 kg of "cathode product" (including approximately 2 kg of transuranics) and 10 Sv/hr 1 m from an IFR fuel assembly containing 90 kg of cathode product (approximately 20 kg of transuranics) produced from 100-day-old spent fuel.[33] When comparing to our results in Table 2, account also must be taken of the fact that the ratio of ^{144}Ce to transuranics would be an order of magnitude lower for IFR spent fuel than for LWR fuel, reducing the associated gamma-radiation field per kg of transuranic material in IFR fuel correspondingly.[34]

Unfortunately, these proliferation-resistance benefits of pyroprocessing are irrelevant to the problem of the already existing and growing U.S. inventory of spent LWR fuel, which, on average, is already more than a decade since discharge and therefore contains an insignificant quantity of ^{144}Ce.

CONCLUSIONS

The radiation doses from transuranics without the lanthanides are more than three orders of magnitude lower than the IAEA's threshold for self-protection.

Inclusion of either of two lanthanide fission products, ^{144}Ce and ^{154}Eu, could increase the dose rate above the self-protection threshold. However, ^{144}Ce has a half-life of only 0.8 years and has already decayed away in all but the most recently discharged spent LWR fuel. ^{154}Eu has a half-life of nine years but is not recycled with the transuranics in the pyroprocessing fuel cycle. It therefore appears that keeping plutonium from aged LWR fuel mixed with other transuranics and with lanthanide fission products other than ^{154}Eu would not make it significantly more self-protecting.

Reprocessing was originally developed to recover plutonium for nuclear weapons from the uranium-metal fuel of plutonium-production reactors. It was then used in Europe and Russia to separate plutonium from higher-burnup oxide power-reactor fuel for startup cores for the plutonium breeder reactors whose large-scale commercialization was originally anticipated in the 1990s. Now, with the indefinite delay of the advent of breeder reactors, the primary rationale has become one of avoiding irreversibly emplacing long-lived transuranic radioisotopes in geological repositories.

In the U.S., however, no irreversible decision on spent-fuel disposition is planned for at least one hundred years. The legislation establishing a national repository requires that it be:

> designed and constructed to permit the retrieval of any spent nuclear fuel placed [there] during an appropriate period of operation of the facility, for any reason pertaining to the public health and safety, or the environment, or for the purpose of permitting the recovery of the economically valuable contents of such spent fuel.[35]

Current DOE plans are to have the period before repository closure last for at least 100 years after emplacement begins.[36]

In this context, it is hard to see an urgent case for separating out transuranics from U.S. spent fuel—especially given that the U.S. and other countries already have to dispose of hundreds of tons of excess plutonium that they previously separated for civilian and weapons purposes.[37]

NOTES AND REFERENCES

1. "U.S. training, aid in Indian A-blast cited" by Don Oberdorfer, *Washington Post*, July 19, 1976, A1; and *Export Reorganization Act of 1976*, Hearings before the U.S. Senate Committee on Government Operations, Jan–March, 1976.

2. "Nonproliferation and export control policy," White House Fact Sheet, Sept. 27, 1993. Another passage states that "the U.S. will seek to eliminate where possible the

accumulation of stockpiles of highly-enriched uranium or plutonium, and to ensure that, where these materials already exist they are subject to the highest standards of safety, security, and international accountability."

3. *National Strategy to Combat Weapons of Mass Destruction*, National Security Presidential Directive17, Dec. 2002, declassified portions available at http://www.fas.org/irp/offdocs/nspd/nspd-17.html

4. He added that "These same technologies offer benefits of enhancing national security by reducing inventories of commercially generated plutonium and enhancing energy independence by recovering the energy value contained in spent nuclear fuel," Statement of Energy Secretary Spencer Abraham on FY 2004 Appropriations, House Committee on Appropriations Subcommittee on Energy and Water Development, March 5, 2003.

5. *Advanced Fuel Cycle Initiative [AFCI]: The Future Path for Advanced Spent Fuel Treatment and Transmutation Research* (Department of Energy, Office of Nuclear Energy, Science and Technology, Report to Congress, 2003), p. I-3. *The National Energy Policy* is the report of Vice President Cheney's 2001 Energy Task force.

6. *Ibid*, p. II-4.

7. "Spent nuclear fuel separations and transmutation criteria for benefit to a geological repository," R. A. Wigeland, T. H. Bauer, T. H. Fanning, and E. E. Morris, Argonne National Laboratory, *Proceedings of Waste Management '04 Conference*, Tucson, AZ, Feb. 29–March 4, 2004. Two thermal limits are assumed. The one that creates the incentive to remove the transuranics is the requirement to maintain a temperature of less than 96°C midway between the tunnels (which are assumed to be spaced 81 m apart) so "that any water flowing downward through the mountain will be able to move through the repository at all times, preventing the retention of a large volume of water above the repository that could flood the repository as it cools." With the removal of the transuranics, a second limit would become constraining: the need to keep the rock immediately surrounding the tunnels below 200°C "to prevent alteration of its crystalline structure." DOE-NE is therefore also exploring the benefits of separating and storing on the surface the 30-year-halflife isotopes, ^{137}Cs and ^{90}Sr, which would otherwise dominate the heat output from the radioactive waste during the period immediately after repository closure. This would allow the storage of the residual radioactive waste from about 40 times as much spent fuel as could be stored in the proposed Yucca Mountain repository in the absence of reprocessing.

8. *AFCI Report to Congress*, Op. Cit., p. II-3.

9. "Designing and demonstration of the UREX+ process using spent nuclear fuel," by G.F. Vandergrift et al., Argonne National Laboratory, presentation at the International conference on advances for future nuclear fuel cycles, Nîmes, France, June 21–24.

10. Advocates of UREX+ point out, however, that the addition of ^{237}Np would have a beneficial impact on the *next* cycle of reprocessing and recycle because neutron capture on ^{237}Np would produce additional quantities of heat-generating ^{238}Pu in the plutonium in the discharged spent fuel.

11. *National Energy Policy* (The White House, May 2001), pp. 5–17; available at www.whitehouse.gov.

12. "Application of the pyrochemical process to recycle of actinides from LWR spent fuel" by C. C. McPheeters, R. D. Pierce, and T. P. Mulcahey, *Progress in Nuclear Energy* 31, 1/2 (1997): 175.

13. Because most of the gamma rays emitted by the transuranic elements have mean-free paths small in comparison with the radius of a 1-kg sphere, the dose rate from

larger spherical masses would increase roughly as the surface area, i.e., the mass to the two-thirds power.

14. *ORIGEN2.1: Isotope Generation and Depletion Code Matrix Exponential Method*, CCC-371 ORIGEN 2.1 (Oak Ridge National Laboratory, Radiation Safety Information Computational Center, August 1996). The PERU50.LIB cross-section files were used to calculate the production rates of actinides and fission products in PWR fuel. The specific power assumed was a constant 37.5 MWt/kgHM (i.e., 3.9 years in core). The ratios for the plutonium isotopes plus ^{241}Am are in good agreement with the results obtained for the same LEU enrichment and burnup in *Plutonium Fuel: An Assessment* (OECD, 1989), Table 9.

15. *Neutron and Gamma-ray Fluence-to-dose Factors* (American Nuclear Society, ANSI/ANS-6.1.1-1991).

16. *MCNP—A General Monte Carlo N-Particle Transport Code, Version 4C*, J. F. Briesmeister, ed, Los Alamos National Laboratory: Los Alamos, NM, LA-13709-M, 2000. We used the Evaluated Nuclear Data File ENDF/B-VI, Brookhaven National Laboratory, July 12, 2001 version. The densities of americium and curium are near 13.5 g/cc. However, the self-shielding is relatively insensitive to density (see below).

17. We give here two examples, ^{241}Am and ^{243}Cm, which α-decay to ^{237}Np and ^{239}Pu respectively. ^{241}Am: The number of decays per kg per hour is 4.7×10^{17}. By far the strongest γ-emission is at 60-keV (36% probability per decay according to the NuDat2 Decay radiation database, Brookhaven National Laboratory, 12/10/04, http://www.nndc.bnl.gov/nudat2/indx_dec.jsp). Before taking into account self-shielding, the flux at a distance of 1 m from the center of the ^{241}Am sphere would be 1.35×10^{12}/cm^2-hr. At 60-keV, the ANS fluence-to-dose conversion factor is 0.4×10^{-12} Sv-cm^2. The dose-rate calculated without taking into account the self-shielding of the plutonium ball would therefore be 0.54 Sv/hr. The mass attenuation length in americium varies from 12.5 to 2 cm^2/g between 50 and 100 keV, *Handbook of Chemistry and Physics, 76th edition* (CRC Press, 1995), pp. 10–287. A power-law interpolation gives a value of 8 cm^2/gm at 60 keV. For an americium density of 13.7 gm/cc (*Ibid*, pp. 4–3) the mean-free path would be $\lambda = 0.009$ cm and the radius of the 1-kg sphere would be R = 2.6 cm. The γ-escape fraction, which is approximately (3λ/4R), would then be about 2.5×10^{-3}, which would reduce the dose rate to 0.0014 Sv/hr, in good agreement with the value in Table 1. ^{243}Cm (using the same references): The most important γ-ray lines are 278 keV (14%) and 228 keV (11%) for 0.25 total emissions per decay with a weighted average energy of 256 keV. Without the self-shielding factor, the flux at 1 meter is therefore 14×10^{12}/cm^2-hr. At 256 keV, the fluence-to-dose conversion factor is 1.33×10^{-12} Sv-cm^2 so the corresponding dose rate would be 18.62 Sv/hr. The γ-ray mass-attenuation coefficient in curium at that energy is about 0.87 cm^2/g. For a density of 13.5, the radius of a 1 kg curium ball is 2.6 cm, the γ-ray mean-free path is 0.085 cm, and the self-shielding factor is 0.025, reducing the dose rate to 0.46 Sv/hr, again in good agreement with the value in Table 1.

18. *The physical protection of nuclear material and nuclear facilities*, International Atomic Energy Agency, INFCIRC/225/Rev.4.

19. *Nonproliferation and arms control assessment of weapons-usable fissile material storage and excess plutonium disposition alternatives* (U.S. Department of Energy, DOE/NN-0007, 1997), pp. 38–39.

20. "Explosive properties of reactor-grade plutonium" by J. Carson Mark, *Science & Global Security* 4: 111.

21. *Nonproliferation and arms control assessment of weapons-usable fissile material storage and excess plutonium disposition alternatives*, op. cit., p. 39. The Department of Energy has also declassified, rather enigmatically, "a definition of pre-initiation-proof weapons (weapon, the yield of which is not sensitive to initiation of the nuclear reaction

at a time earlier than the planned time)," *'Drawing back the curtain of secrecy:' Restricted data declassification decisions, 1946 to the present*, RDD-3, U.S. Department of Energy, Office of Declassification, Jan. 1, 1996, V.C.2.n, p. 96.

22. *IAEA Safeguards Glossary, 2001 edition*, p. 23

23. The laminar convective heat transfer coefficient for a sphere in air is $1.32[\Delta T/R]^{0.25}$ Watts/m^2-°K, where R is the radius of the sphere and ΔT is the temperature difference between it and the ambient air, *2001 ASHRAE Handbook: Fundamentals*, American Society of Heating, Refrigerating and Air-Conditioning, 2001, p. 3.12, Table 5. Radiation loss from a surface with a hemispherical emittance ε is $\varepsilon\sigma T^4$, where we take $\varepsilon = 0.95$ for the surface of the sphere (assumed to be painted black) and 0.9 for the building surfaces which are radiating back at the sphere (assumed to be at a temperature of 20°C). $\sigma = 5.67 \times 10^{-8}$ Watts/m^2 $-^\circ$ K^4 is the Stefan-Boltzman constant, ibid, p. 3.8.

24. *Explosives Science* by R. N. Rogers and J. L. Rogers, Los Alamos, NM, http://home.att.net/~rnrogers/, undated.

25. In the Nagasaki bomb design, the plutonium sphere and concentric uranium and aluminum spheres had radii of 4.6, 11.4 and 23.5 cm, respectively, *Atom Bombs* by John Coster-Mullen, self-published, 2005. The heat capacities of uranium and aluminum metal at 25°C are 2.2 and 2.4 j/(cc-°C), respectively. With a heat input of 160 Watts, the heat up rate of the uranium and aluminum would be about 4.5°C/hr.

26. *The Making of the Atomic Bomb* by Richard Rhodes, Simon and Schuster-Touchstone1988, pp. 659–661.

27. The bare critical mass of weapon-grade uranium containing 93.71% ^{235}U is 49 kg, *Critical dimensions of systems containing* ^{235}U, ^{239}Pu, and ^{233}U by H. C. Paxton and N. L. Pruvost, Los Alamos National Laboratory, LA-10860-MS, 1987.

28. *Nuclear Chemical Engineering*, *2nd edition*, M. Benedict, T. H. Pigford, and H. W. Levi (McGraw-Hill, 1981), Table 8.1.

29. The fission yield of ^{144}Ce is 0.055 per fission from ^{235}U and 0.037 from ^{239}Pu, for an average of 0.046, T. R. England and B. F. Rider, Los Alamos National Laboratory, LA-UR-94-3106; ENDF-349 (1993), http://ie.lbl.gov/fission.html/. Ignoring transmutation and assuming a 4-year period in the reactor, about 27% would remain at discharge or 0.012 per fission. There are 0.24 transuranics per fission. Therefore, per kg of transuranics, there should be 30 grams of Ce-144 at discharge, in good agreement with Table 2. The dominant ^{144}Ce γ emission (11% of decays) has an energy of 0.134 MeV. Without the self-shielding factor, at 1 m, the flux from 32 grams would be 12×10^{12}/hr-cm^2. The ANS γ-ray fluence-to-dose factor for that energy is 0.7×10^{-12} Sv-cm^2, so the unshielded dose rate would be 8.4 Sv/hr. The mass-attenuation coefficient of plutonium at that energy is about 0.19 cm^2/g. For a density of 19.84 grams/cc, the mean-free path is $\lambda = 0.027$ cm and the radius of a 1 kg plutonium sphere is R = 2.3 cm, so the self-shielding factor ($3\lambda/4R$) is 0.087, giving a dose rate of 0.07 Sv/hr. ^{144}Pr emits γ rays with energies of 2.2 MeV (0.7% per decay), 1.5 MeV (0.28%), and 0.70 MeV (1.3%). The corresponding ANS fluence-to-dose factors are 8.16, 6.24, and 3.41×10^{-12} Sv-cm^2, respectively. In the absence of self-shielding, the corresponding γ-ray doses 1 m from 1.37×10^{-3} grams of ^{144}Pr would be 6.43, 1.97, and 4.99 Sv/hr respectively, for a total of 13.4 Sv/hr. The mass-attenuation coefficients in plutonium of the three γ rays are 0.050, 0.067, and 0.134 cm^2/gram. The corresponding γ mean-free paths in plutonium are $\lambda = 1.01$, 0.75 and 0.38 cm. The corresponding self-shielding factors ($3\lambda/4R$) are 0.33, 0.24, and 0.124, which reduces the doses to 2.10, 0.47 and 0.62 Sv, respectively for a summed dose from ^{144}Pr of 3.2 Sv/hr, corresponding to a weighted average self-shielding factor for the three γ lines of 0.24. The discrepancy with the MCNP result in Table 2 (a factor of 0.4) is larger than for the other isotopes that were hand calculated. Investigating further, we found that the dose from the 2.2 MeV line is in agreement but that MCNP obtains higher doses

from the lower lines and then considers other lines that increase the dose by a further 50%.

30. "Application of the pyrochemical process to recycle of actinides from LWR spent fuel," op. cit.

31. Estimates of the fraction of ^{144}Ce that would accompany the transuranics range from 10–25%, "Physics studies of higher actinide consumption in an LMR" by R. N. Hill, D. C. Wade, E. K. Fujita, and H. Khalil, Proceedings of the International Conference on the Physics of Reactors, Marseille, France, 1990, I-83.

32. "Nonproliferation and Safeguards Aspects of the IFR" by W. H. Hannum, D. C. Wade, H. F. McFarlane, and R. N. Hill, *Progress in Nuclear Energy*, 31, 1/2 (1997): 203.

33. "Nonproliferation and Safeguards Aspects of the IFR," op. cit, Figure 3. The treatment of self-shielding in this article is confusing because, it is stated in the text that a γ-radiation dose can be estimated by assuming that one Watt of γ power translates into a dose rate of 1 Sv/hr at 1 m. This is approximately true for an unshielded dose from a ^{144}Pr source. In the absence of self-shielding, an isotropic point source of 1 Watt of gamma power translates into a flux of 1.8×10^{11}(1/E) photons/cm^2-hr at 1 m, where E is the average energy of the γs in MeV. ^{144}Pr emits γ rays with an average energy of 1.24 MeV. The fluence-to-dose factor at that energy is about 5.4×10^{-12} Sv-cm^2, *Neutron and gamma-ray fluence-to-dose factors*, op. cit. This results in an estimate of a dose of 0.78 Sv/hr. The nonlinearity with quantity of transuranics of the doses quoted in the Argonne paper for the cathode product and IFR fuel assembly suggests, however, that self-shielding has been taken into account.

34. The fast fission yield of ^{144}Ce from ^{239}Pu is 0.037 per fission—about 80% the average of that from thermal fission of ^{235}U and ^{239}Pu (see footnote above, and same reference). Spent IFR fuel contains about 22% transuranics vs. about 1% in spent LWR fuel and is assumed to contain10% fission products vs. about 5% for spent LWR fuel, "Nonproliferation and Safeguards Aspects of the IFR," op. cit, Tables 1 and 2.

35. National Waste Policy Act of 1982, Sec. 122.

36. *Final Environmental Impact Statement for a Geologic Repository for the Disposal of Spent Nuclear Fuel and High-Level Radio active Waste at Yucca Mountain, Nye County, Nevada, [Final Yucca Mt EIS]* DOE/EIS-0250, 2002, Summary, http://www.ocrwm.doe.gov/documents/feis_2/summary/indexsum.htm/.

37. As of the end of 2003, more than 230 tons of separated civilian plutonium had been declared to the IAEA, David Albright and Kimberley Kramer, "Separated Civil Plutonium Inventories: Current Status and Future Directions," Revised July 8, 2005, http://www.isis-online.org/global_stocks/separated_civil_pu.pdf, Table 1. Of this, about half was owned by the U.K. and Russia, neither of which has a disposal plan. The U.S. and Russia have each also declared excess 34 tons of weapon plutonium and are likely to increase these declarations in the future.