Science & Global Security, Volume 9 pp 1-32

0892-9882/01 $12.00 + .00

U-232 and the Proliferation-Resistance of U-233 in Spent Fuel

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The factors influencing the level of U-232 contamination in U-233 are examined for heavy-water-moderated, light-water-moderated and liquid-metal cooled fast breeder reactors fueled with natural or low-enriched uranium and containing thorium mixed with the uranium or in separate target channels. U-232 decays with a 69-year half-life through 1.9-year half-life Th-228 to Tl-208, which emits a 2.6 MeV gamma ray upon decay.

We find that pressurized light-water-reactors fueled with LEU-thorium fuel at high burnup (70 MWd/kg) produce U-233 with U-232 contamination levels of about 0.4 percent. At this contamination level, a 5 kg sphere of U-233 would produce a gamma-ray dose rate of 13 and 38 rem/hr at 1 meter one and ten years after chemical purification respectively. The associated plutonium contains 7.5 percent of the undesirable heat-generating 88-year half-life isotope Pu-238.

However, just as it is possible to produce weapon-grade plutonium in low-burnup fuel, it is also practical to use heavy-water reactors to produce U-233 containing only a few ppm of U-232 if the thorium is segregated in "target" channels and discharged a few times more frequently than the natural-uranium "driver" fuel. The dose rate from a 5-kg solid sphere of U-233 containing 5 ppm U-232 could be reduced by a further factor of 30, to about 2 mrem/hr, with a close-fitting lead sphere weighing about 100 kg.

Thus the proliferation resistance of thorium fuel cycles depends very much upon how they are implemented.

The original version of this manuscript was received by *Science & Global Security* on 15 March, 2001.

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INTRODUCTION

Uranium-233 is, like plutonium-239, a long-lived fissile isotope produced in reactors by single-neutron capture in a naturally-occurring abundant fertile isotope (see Figure 1). The fast critical mass of U-233 is almost identical to that for Pu-239 and the spontaneous fission rate is much lower, reducing to negligible levels the problem of a spontaneous fission neutron prematurely initiating the chain reaction -- even in a "gun-type" design such as used for the U-235 Hiroshima bomb (see Table 1). Why then has plutonium been used as the standard fissile material in the "pits" of modern nuclear weapons while U-233 has not? This question is not just of historical interest, since there is increasing interest in U-233-thorium fuel cycles.

Table 1: Comparison of some nuclear characteristics of U-233, U-235, and Pu-239

Property	**Uranium-233**	**Uranium-235**	**Plutonium-239**
Half-life (years)	1.6×10^5	7.0×10^8	2.4×10^4
Fertile isotype (continental crust mass abundance[a]) and thermal neutron absorption cross-section	TH-232 (9.6ppm) 7.4 barns (10^{-24} cm^2)	U^{235} is derived from natural uranium.	U-238 (2.7 ppm) (99.3% in U_N) 2.7 barns
Critical Mass (kg) (reflector)[b]	8.4 (98.3% U^{233}) (3.7 cm Be)	21 (93.5% U^{235}) (5.1 cm Be)	7.5 (4.9% Pu^{240}) (4.2 cm Be)
Neutrons released per neutron absorbed (energy of neutron causing fission)[c]	2.5 (1 MeV) 2.28 (0.025 eV)	2.3 2.07	2.9 2.11
Spontaneous fission rate (sec-kg)$^{-1}$	0.5	0.6 (for 1% U^{234} and 5.5% U^{238}	2.5×10^4 (for 6% Pu^{240}
Decay heat (W/kg)	0.3	10^{-4}	2.4 (6% Pu^{240})
Delayed neutron fraction[d]	0.00266	0.0065	0.00212

a. *Handbook of Chemistry and Physics, 76th edition* (1995-6): 14-11.
b. *Critical Dimensions of Systems Containing U-235, Pu-239, and U-233* (LA-10860-MS, Los Alamos National Laboratory, 1986 revision). Plutonium, 15.6 gm/cc; U-233, 18.6 gm/cc.
c. A.M. Perry and A.M. Weinberg, "Thermal Breeder Reactors," Annual Review of Nuclear and Particle Science, 22 (1972): 317-354.
d. *Thorium based fuel options for the generation of electricity: Developments in the 1990s* (Vienna, IAEA-TECDOC-1155, May 2000): 9.

$$n + {}^{238}U \longrightarrow {}^{239}U \xrightarrow[24\ m]{\beta^-} {}^{239}Np \xrightarrow[2.4\ d]{\beta^-} {}^{239}Pu \text{ (24,000 years)}$$

$$n + {}^{232}Th \longrightarrow {}^{233}Th \xrightarrow[22\ m]{\beta^-} {}^{233}Pa \xrightarrow[27.4\ d]{\beta^-} {}^{233}U \text{ (160,000 years)}$$

Figure 1: Production of Pu-239 and U-233.

In this paper we examine: how U-233 can be produced in existing reactor types; its attractions as a reactor fuel; the determinants of the co-production of U^{232}, one of whose decay products emits hard gamma rays; and the influence of that isotope at various contamination levels on the weapons-usability of U-233. Our findings are summarized in the body of the paper. The calculational tools are described in the Appendices.

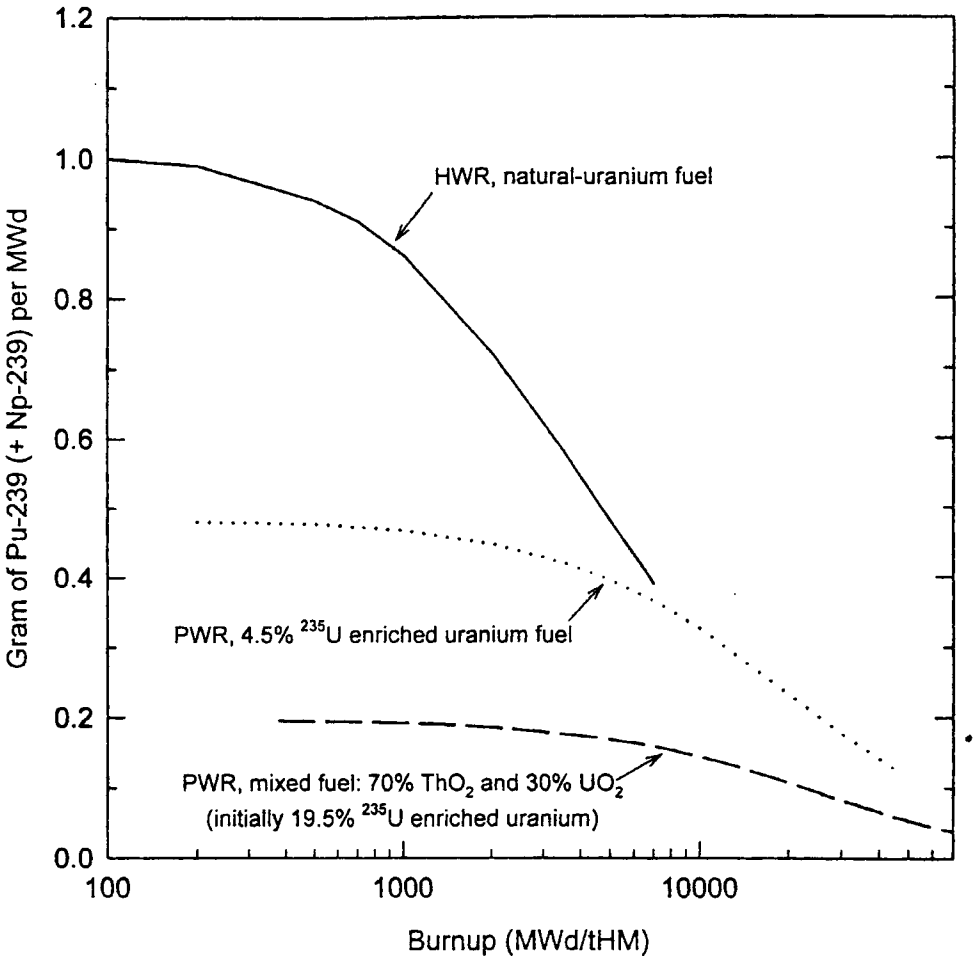

Figure 2: Net production rate of Pu-239 per Megawatt-day (MWd) of fission energy released as a function of burnup in HWR and pressurized light-water reactor (PWR) fuel.

U-233 Production

One of the most important reasons why plutonium was chosen over U-233 as a weapons material is that first-generation plutonium-production reactors were fueled by natural uranium, which contains almost as large a fraction of neutron-absorbing fertile material (U-238) as is possible consistent with a reactor achieving criticality. In a natural-uranium fueled reactor, such as the Canadian heavy-water-moderated (HWR) reactor type, Pu-239 is produced by neutron absorption in U-238 at a rate of about one gram of plutonium per thermal

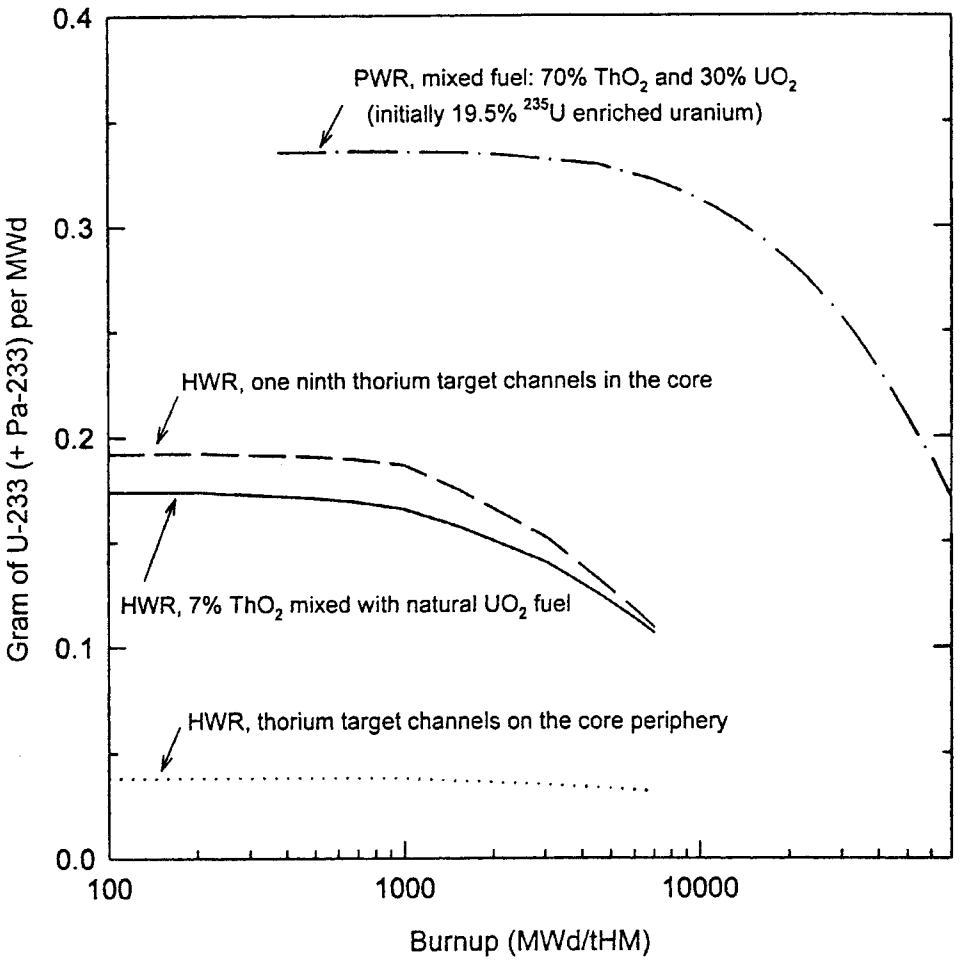

Figure 3: Net production rate of U-233 as a function of burnup of driver fuel per MWd in a PWR and in a natural-uranium-fueled HWR for thorium mixed in the fuel, or in separate channels (one out of nine), or in channels on the periphery of the core.

megawatt-day (MWd) of fission energy release at low U-235 "burnups," (see Figure 2).[1] Approximately one MWd is released by the fission of one gram of fissile material. After taking into account the neutron requirements for maintaining a steady chain reaction, there is about one excess neutron available per fission and virtually all of these neutrons are absorbed by U-238.[2]

Production of U-233 requires the addition of the fertile material Th-232. If the fuel is natural uranium, only a relatively small percentage of thorium can be added before it becomes impossible to sustain a chain reaction. We estimate that about 7 percent thorium oxide can be added to HWR fuel before the

achievable burnup is reduced from 7000 to 1000 MWd/t (thermal megawatt-days per ton-heavy metal). Because the thermal-neutron absorption cross-section of Th-232 is almost 3 times larger than that of U-238, this concentration of thorium would yield about 0.2 grams of U-233 per MWd at burnups lower than 1000 MWd/t (see Figure 3). Thus most of the fissile material produced in the core would still be plutonium.

In all the figures in this article, we include in the production of Pu-239 and U-233, the production of their short-lived precursors, Np-239 (2.4-day half-life) and Pa-233 (27-day half-life) respectively.

For a country with uranium-enrichment capabilities, the balance between plutonium and U-233 production could be shifted almost all the way toward U-233 by fueling production reactors with highly-enriched uranium. Indeed the U.S. produced much of its weapons plutonium in the Savannah River heavy-water-moderated production reactors, using highly-enriched uranium fuel and depleted uranium targets in mixed-lattice arrangements.[3]

U-232 Radiation Hazard

A second problem with U-233 as a fissile material for either weapons or reactor fuel is that it contains an admixture of U-232, whose decay chain produces penetrating gamma rays. The decay chain of U-232 is shown in Figure 4. The most important gamma emitter, accounting for about 85 percent of the total dose from U-232 after 2 years, is Tl-208, which emits a 2.6-MeV gamma ray when it decays (see Appendix C). For plutonium containing a significant admixture of 14.4-year half-life Pu-241, the most important source of gamma-ray irradiation from is its 433-year half-life decay product, Am-241, which emits low-energy (< 0.1 MeV) gamma rays. These gamma rays do not represent a significant occupational hazard for weapon-grade plutonium (0.36% Pu-241) but their dose becomes more significant for “reactor-grade” plutonium, which contains on the order of 10 percent Pu-241. Thus both U-233 contaminated with U-232 and reactor-grade plutonium are made less desirable as weapons materials by virtue of the fact that their gamma emissions bring with them the potential for significant radiation doses or shielding requirements for workers involved in nuclear weapons production and for military personnel handling nuclear weapons.

Figure 5a shows the calculated buildup with time of the gamma dose rate 0.5 meters (a typical working distance for glove-box operations) from 5-kg spheres of freshly separated U-233 containing 0, 1 and 5 ppm of U-232. It will be seen that the dose rate from pure U-233 is of the same order as that from

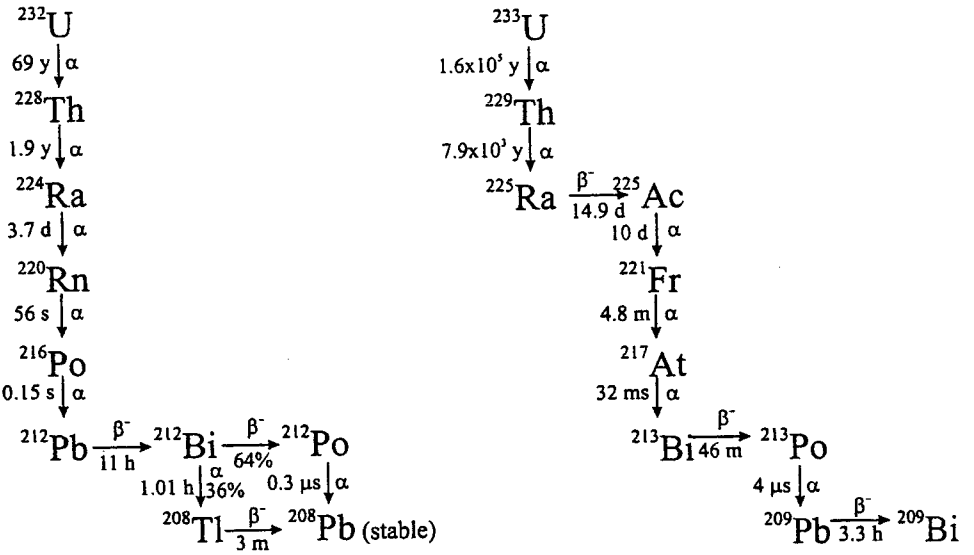

Figure 4: Decay chains of U-232 and U-233.

weapon-grade plutonium. For U-233 containing U-232, the buildup in dose rate with time reflects the in-growth of Th-228, which has a half-life of 1.9 years. After this in-growth, the dose rate from U-233 containing 1 ppm U-232 is about the same as reactor-grade plutonium after a large fraction of its 14.1-year half-life. Pu-241 (initially 9.1 % of the plutonium)[4] has decayed to Am-241.

Figure 5b shows the effectiveness of lead shielding in reducing the dose from 5 kg spheres of reactor-grade plutonium and U-233 as a function of the weight of lead in a close-fitting shell. It will be seen that the lower-energy

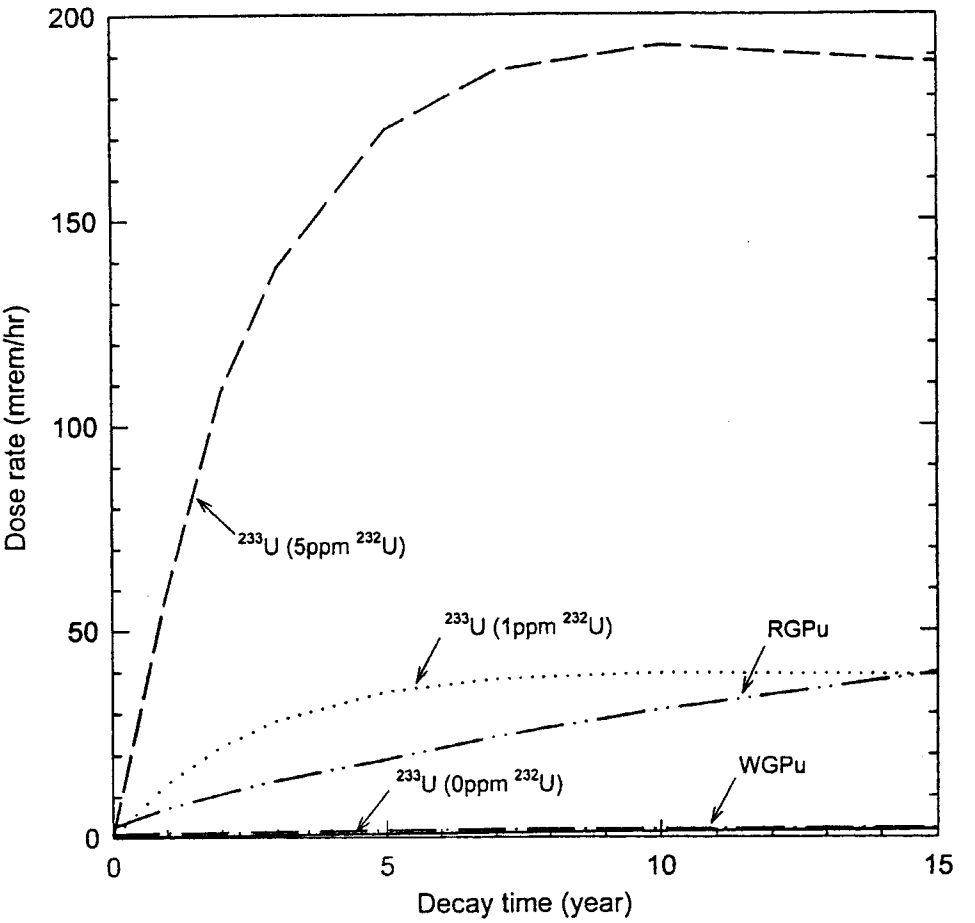

Figure 5a: Radiation-dose-rate buildup at 0.5 m from 5-kg spheres of U-233 and Pu-239 for different admixtures of U-232 and higher plutonium isotopes, respectively.

gamma rays from the plutonium are easily shielded. Shielding the neutron dose from spontaneous fission of the even-numbered plutonium isotopes in reactor-grade plutonium would require a relatively thick layer of neutron moderator containing hydrogen (e.g. plastic) followed by a layer of neutron absorbing material and then additional shielding from the gamma-rays produced when the neutrons are captured.

Occupational radiation doses are currently limited to 5 rem/yr in the US.[5] A worker could be 0.5 meters from an unshielded 5-kg sphere of 1-year-separated weapon-grade plutonium (dose rate, 1.3 mrem/hr) for almost 3800 hours

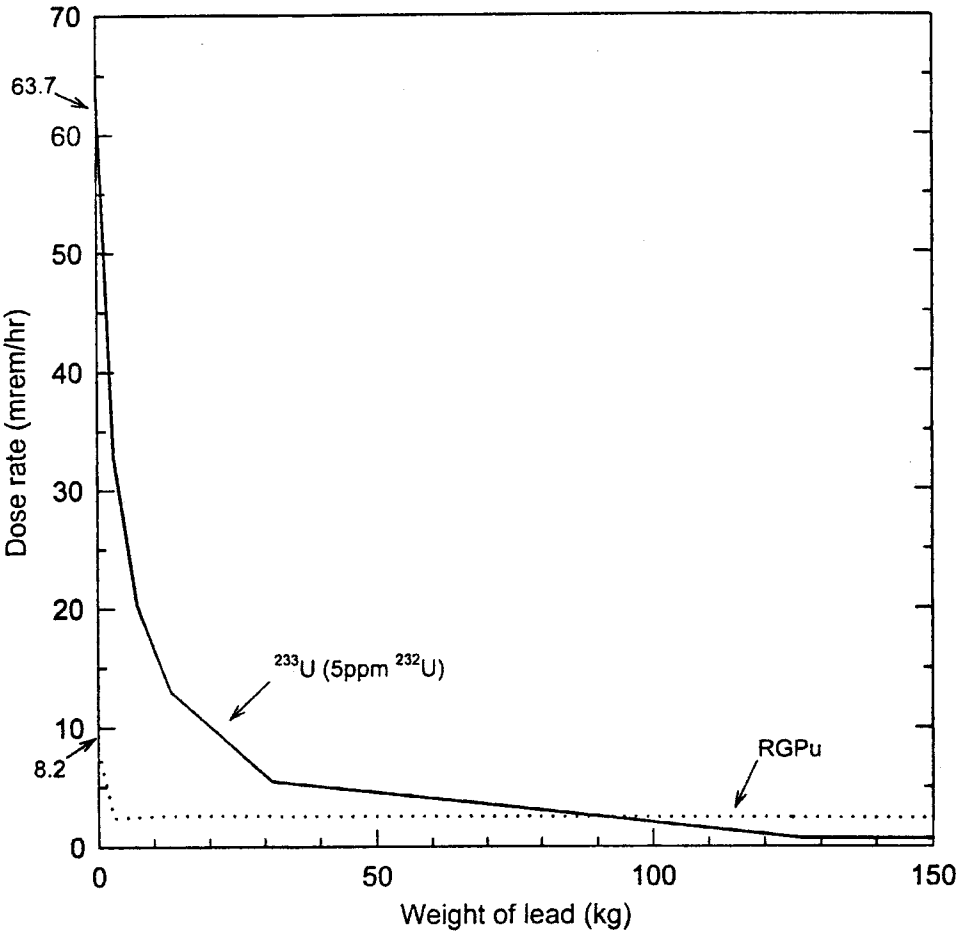

Figure 5b: Radiation-dose-rate attenuation for one-year-old separated reactor-grade plutonium and U-233 containing 5 ppm U-232 as a function of the weight of a close-fitting hollow-sphere lead shield.

before reaching that dose limit. After in-growth of Am-241, the dose rate from a sphere of reactor-grade plutonium one year after separation would be about 8.2 mrem/hr, limiting the worker to about 610 contact hours per year. The situation is about the same with U-233 containing 1 ppm U-232 after in-growth of Tl-208. For 1-year-separated U-233 containing 5 ppm U-232, a worker could only be allowed about 80 contact hours. Thus maximizing the contamination level of U-232 in U-233 would make it both significantly more difficult to fabricate and make it more detectable because of the difficulty of shielding the 2.6 MeV gamma ray. However, it would require a level of 2.4 percent U-

232 before the U-233 would satisfy the IAEA's standard for reduced physical-protection requirements (>100 rem/hr at 1 meter).[6]

Table 2: Unshielded working hours required to accumulate a 5 rem dose (5 kg sphere of metal at 0.5 m one year after separation)

Metal	Dose Rate (rem/hr)	Hours
Weapon-grade plutonium	0.0013	3800
Reactor-grade plutonium	0.0082	610
U-233 containing 1ppm U-232	0.013	380
U-233 containing 5ppm U-232	0.059	80
U-233 containing 100 ppm U-232	1.27	4
U-233 containing 1 percent U-232	127	0.04

India's Department of Atomic Energy (DAE) has been concerned about the occupational hazards associated with the fabrication of fuel containing U-233. Its long-term ambition is to cleanse U-233 down to "a few ppm" U-232 using laser isotope purification.[7] In the meantime, a 1993 article from the Bhabba Atomic Research Center in Bombay reported a 6.7 person-rem summed dose incurred by workers fabricating a research-reactor core containing 0.6 kg "clean" U-233 containing 3 ppm U-232.[8]

Interest in U-233 as a Reactor Fuel

One reason for interest in U-233 as a reactor fuel is the superior conversion ratios C_R that can be achieved with it in slow-neutron reactors.[9] It will be seen from Table 1 that about 0.2 more neutrons are produced on average per "thermal" (20 ^oC or 0.025 eV) neutron absorbed on U-233 than for absorption in Pu-239. This difference increases with neutron energy to about 0.4 at 0.1 eV neutron energy and to about 0.6 at 0.3 eV. These are important differences because the amount of fissile material required per megawatt-day of fission energy released from closed fuel cycles is proportional to $1\text{-}C_R$. In fact, the Th-232/U-233 fuel cycle can have $C_R > 1$, i.e. be a net "breeder" of fissile material in thermal-neutron reactors if the use of neutron "poisons" to control excess reactivity is minimized by use of continuous fueling or geometry control of reactivity.

There was a great deal of interest in breeder reactors from the 1940s through the 1970s. During this period it was believed that world nuclear-power capacity would rapidly outgrow the ability of the world's high-grade

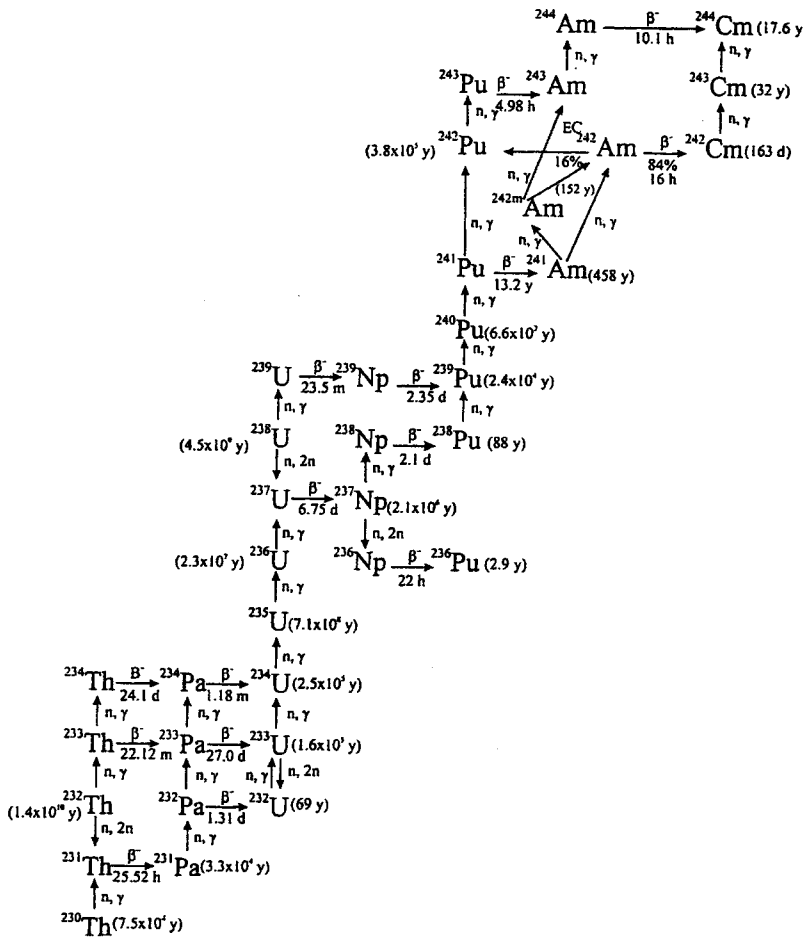

Figure 6: Products of multiple-neutron captures on Th-230, Th-232, U-235, and U-238.

uranium resources to support reactors fueled by natural or low-enriched uranium and operating on "once-through" fuel cycles. Most attention was focused on fast-neutron breeder reactors based on a "closed" U-238/Pu-239 fuel cycle (i.e. one involving the recycle of plutonium). But detailed studies, experiments, and even demonstrations were carried out with various types of thermal-neutron breeder reactors operating on a closed Th-232/U-233 fuel cycle, including: heavy-water reactors,[10] light-water reactors,[11] and molten-salt reactors.[12]

Interest in breeder reactors has waned because world nuclear-power

capacity is an order of magnitude lower than projected in the mid-1970s; reprocessing and plutonium fuel fabrication costs are an order of magnitude higher; and uranium costs are an order of magnitude lower. However, interest in the thorium fuel cycle continues in India, because of its relatively small uranium reserves, large thorium resources, and the unwillingness of uranium exporters to sell it uranium because it is not a party to the Nonproliferation Treaty. Indeed, India's nuclear establishment continues to adhere to the 3-stage plan of nuclear-energy development laid out in the 1950s by its founder, Homi Bhabba. The first stage involves the use of HWRs fueled by natural uranium and light-water reactors fueled by low-enriched uranium. In the second stage, plutonium extracted from the spent fuel of these reactors is be used as startup fuel for liquid-sodium-cooled fast breeder reactors. In the third stage, U-233 produced by neutron capture in the thorium blankets of these breeder reactors would be mixed with thorium and used to start up heavy-water and perhaps also high-temperature gas-cooled reactors operating on a closed Th-232/U-233 fuel cycle.[13]

Recently, there has also been a revival of interest in thorium in the U.S. and Western Europe because it can be used to increase the achievable burn-ups in light-water-reactors operating on a once-through fuel cycle and also reduce the quantity of weapons-usable transuranic elements in radioactive waste.[14] Five successive neutron captures are required before Np-237 is produced from Th-232 whereas a single neutron capture on U-238 produces Pu-239 (see Figure 6).[15] Proposals have therefore been brought forward for light-water-reactor designs in which thorium largely replaces U-238[16] and for accelerator-driven fast-neutron sub-critical reactors that would produce U-233 out of thorium.[17]

Isotopic "Denaturing" of U-233 for weapons use

According to the IAEA, the enrichment boundary below which enriched uranium is not directly usable to make fission explosives is 20 percent U-235. Thus dilution by U-238 "denatures" U-235 for weapons purposes. There is no comparable isotopic dilutant for plutonium.[18] However, U-238 is available in abundance in natural and depleted uranium to denature U-233. Figure 7 shows the reflected critical mass of U-233 and U-235 mixtures with U-238 as a function of percentage of the fissile isotope.[19] It will be seen that the critical mass of a sphere of uranium 20-percent enriched in U-235 and surrounded by a 4-cm thick layer of beryllium is about 400 kg. A U-233/U-238 mixture has a corresponding critical mass when the U-233 percentage is approximately 12

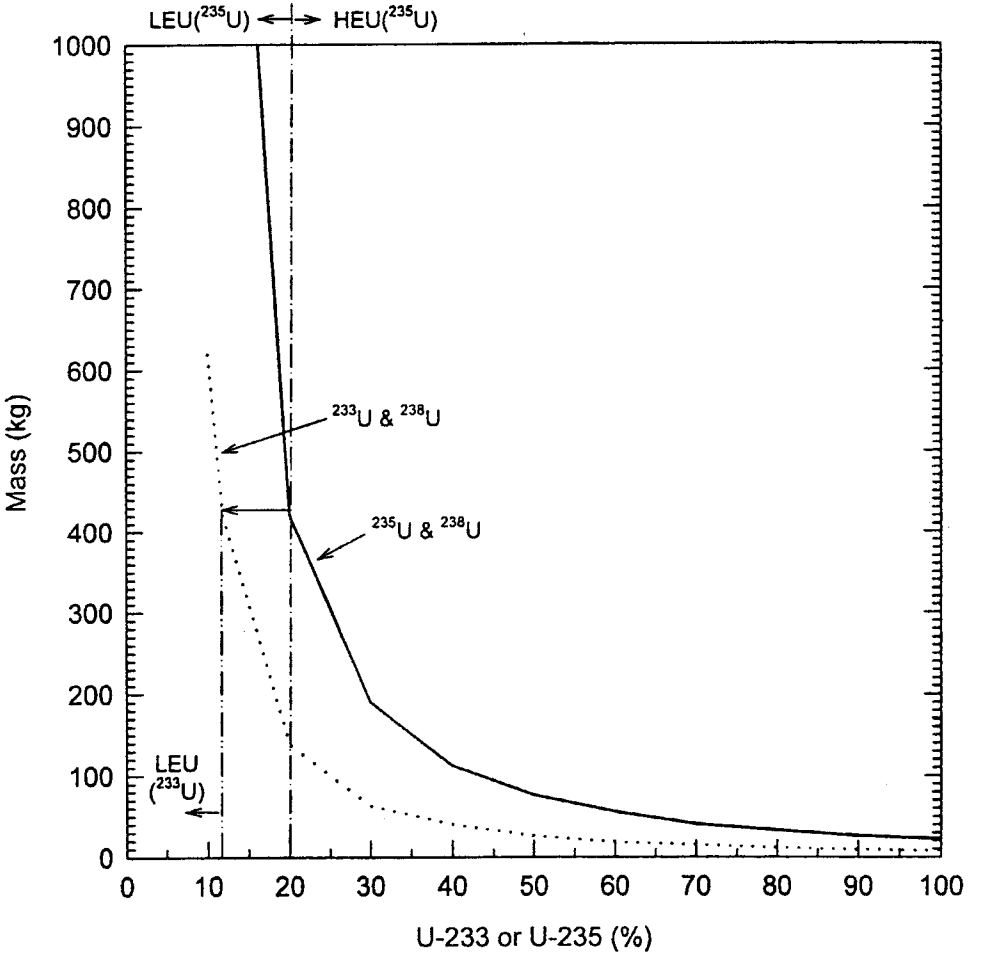

Figure 7: Reflected critical masses as a function of percentage of U-233 or U-235 in isotopic mixture with U-238.

percent.

Uranium enriched to just under 20 percent in U-235 may not be directly useable to make a nuclear explosive. However, it requires only about one quarter as much enrichment work to enrich to weapon grade (90 percent U-235) as from 4.5% enriched uranium.[20]

Determinants of U-232 concentration in U-233. U-232 is produced from Th-232 via two of the reaction chains shown in Figure 6. Each of these chains involves a neutron-absorption (n,γ) reaction and a reaction in which an incoming neutron knocks two neutrons out of a target nucleus [(n,2n) reac-

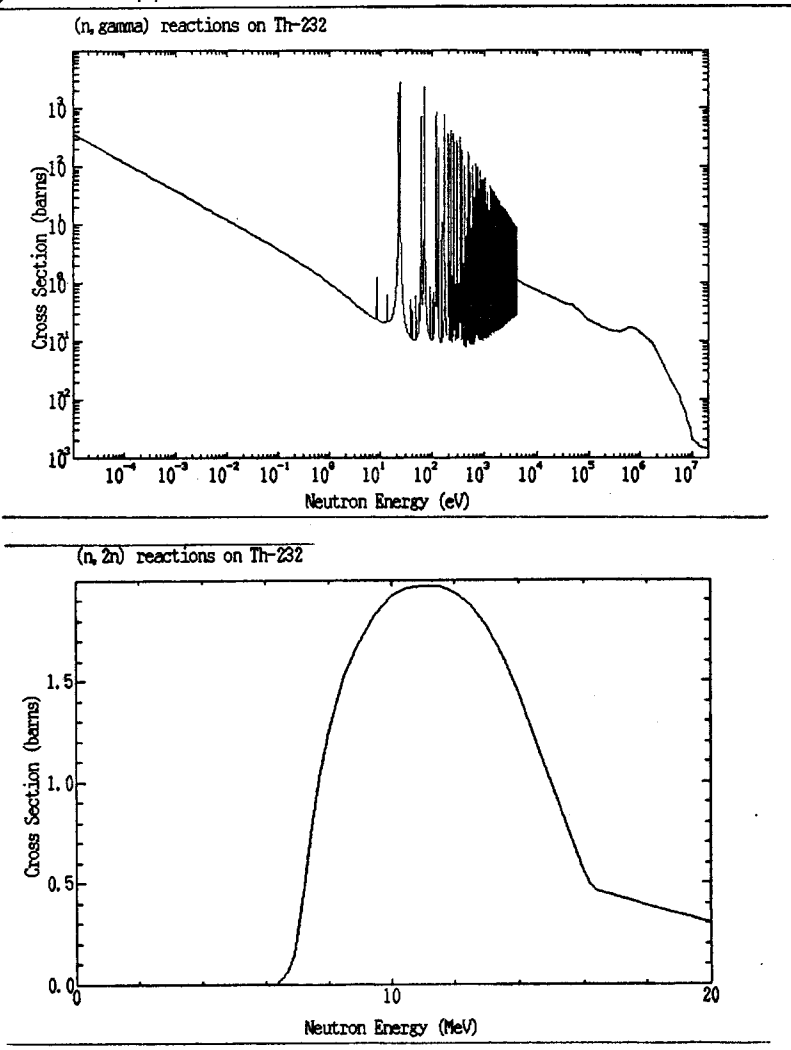

Figure 8: Cross-sections for neutron absorption: a) (n, gamma) and b) (n,2n) reactions on Th-232.

tion]. U-232 can also be produced by two successive single neutron captures starting with naturally-occurring Th-230. Thorium-230 is a decay product of U-234, which is in turn a decay product of U-238, is in secular equilibrium at a concentration of about 17 ppm in natural uranium. Minimizing U-232 production therefore requires naturally thorium that is minimally contaminated with Th-230 from intermixed or nearby natural uranium. In the calculations described below, we have assumed zero Th-230 contamination and have tested the sensitivity of the results to a contamination level of 1 ppm.

The threshold neutron energy required for the (n,2n) reactions involved in

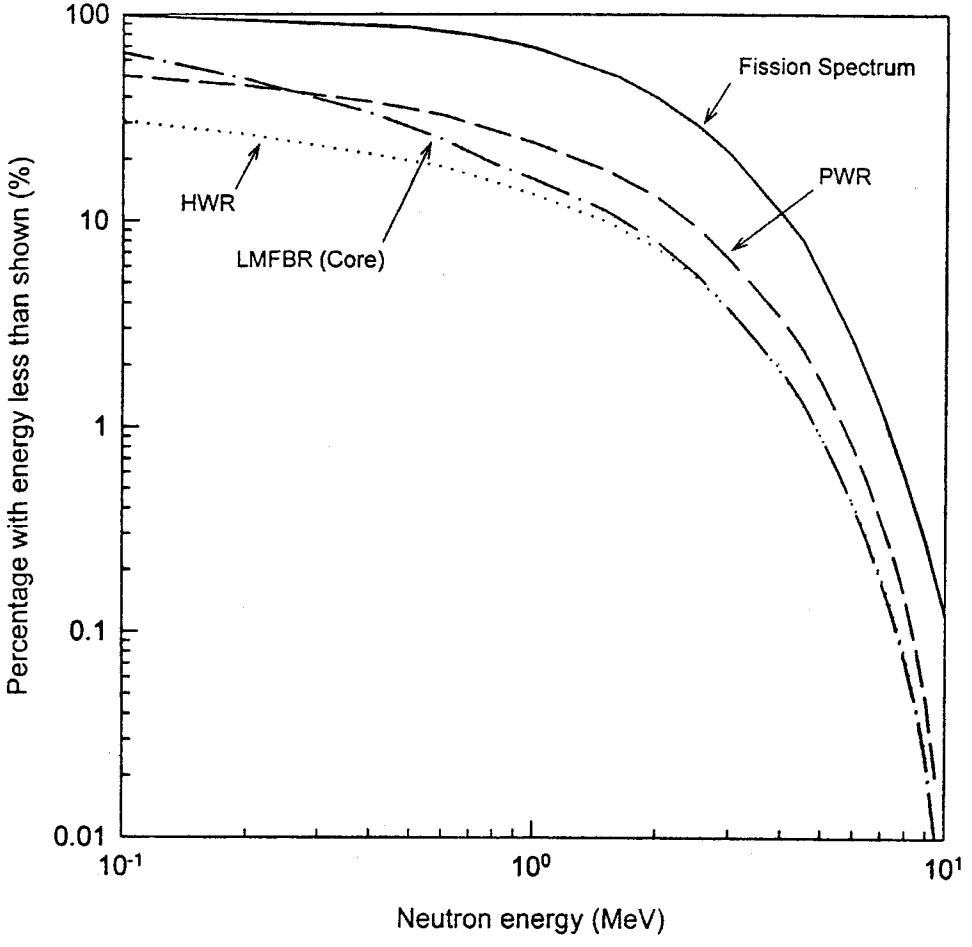

Figure 9a: Neutron-energy spectra: a) Fission spectrum compared with spectra in fuel of an HWR and cores of an LMFBR and PWR.

U-232 production is around 6 MeV.[21] Such energies are found only in the high-energy tail of the fission spectrum. (Figure 8 shows the cross-section for neutron-capture [n, γ] and [n, 2n] reactions on Th-232.[22]) The fission-spectrum average cross-sections are 14.46 mb for the reaction $n + Th232 \rightarrow Th231 + 2n$ and 4.08 mb for the reaction $n + U233 \rightarrow U232 + 2n$.[23] The development of the U-232/ U-233 concentration ratio in thorium therefore depends upon the fraction of the neutron fluence above 6 MeV in the thorium target material.

Figure 9a and Table 3 show a fission neutron-energy spectrum and compare it with the neutron energy spectra in the fuel of HWR, PWR and LMFBR

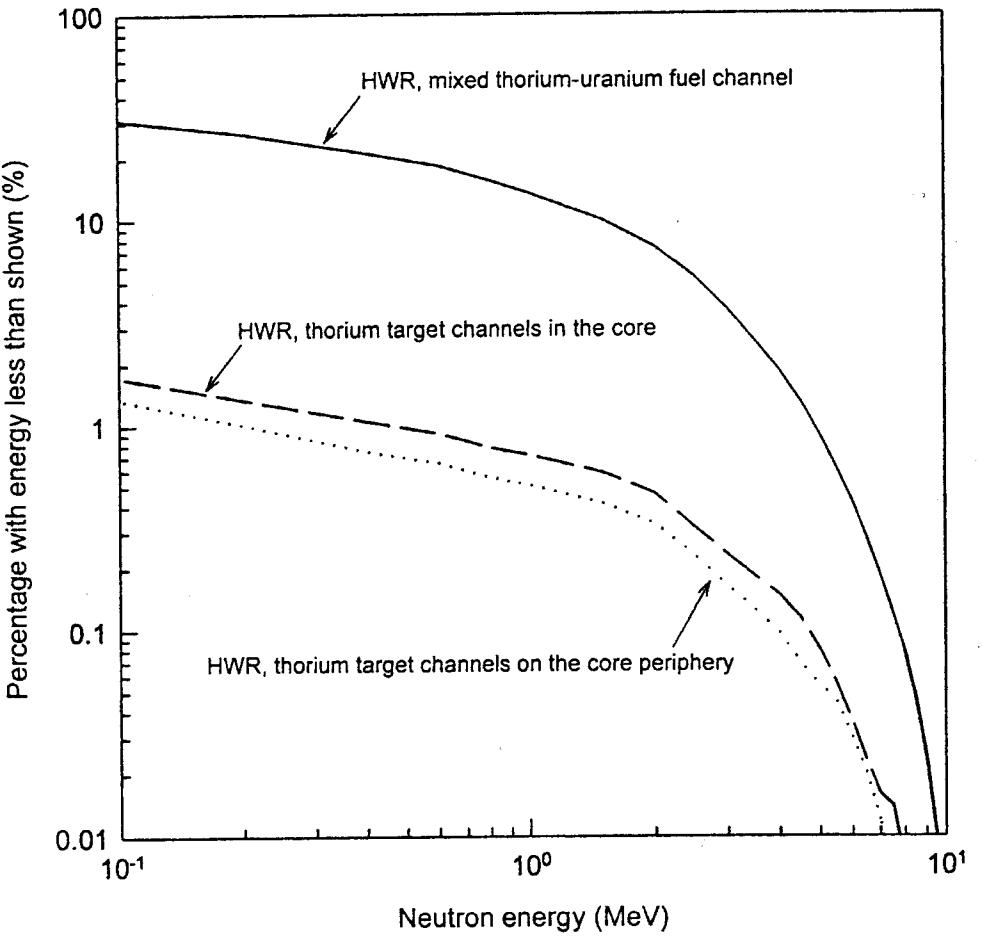

Figure 9b: Neutron-energy spectrum in HWR fuel compared with those in mixed thorium-uranium fuel channel and thorium "target" channels in the core and on the core periphery.

reactors.[24] The portions of the neutron fluence above 6 MeV are reduced respectively by factors of about 0.15, 0.3 and 0.15 relative to the fission spectrum. The high-energy fluence is still less in a core location away from the fuel. Figure 9b shows the neutron spectrum in HWR channels filled with thorium "target" assemblies inside and in the periphery of an HWR core compared with the neutron spectrum in a fuel channel. In both cases, the flux in the high-energy tail is reduced by a factor of about 0.01 relative to the fission spectrum. Figure 9c shows the neutron spectrum in an LMFBR radial tho-

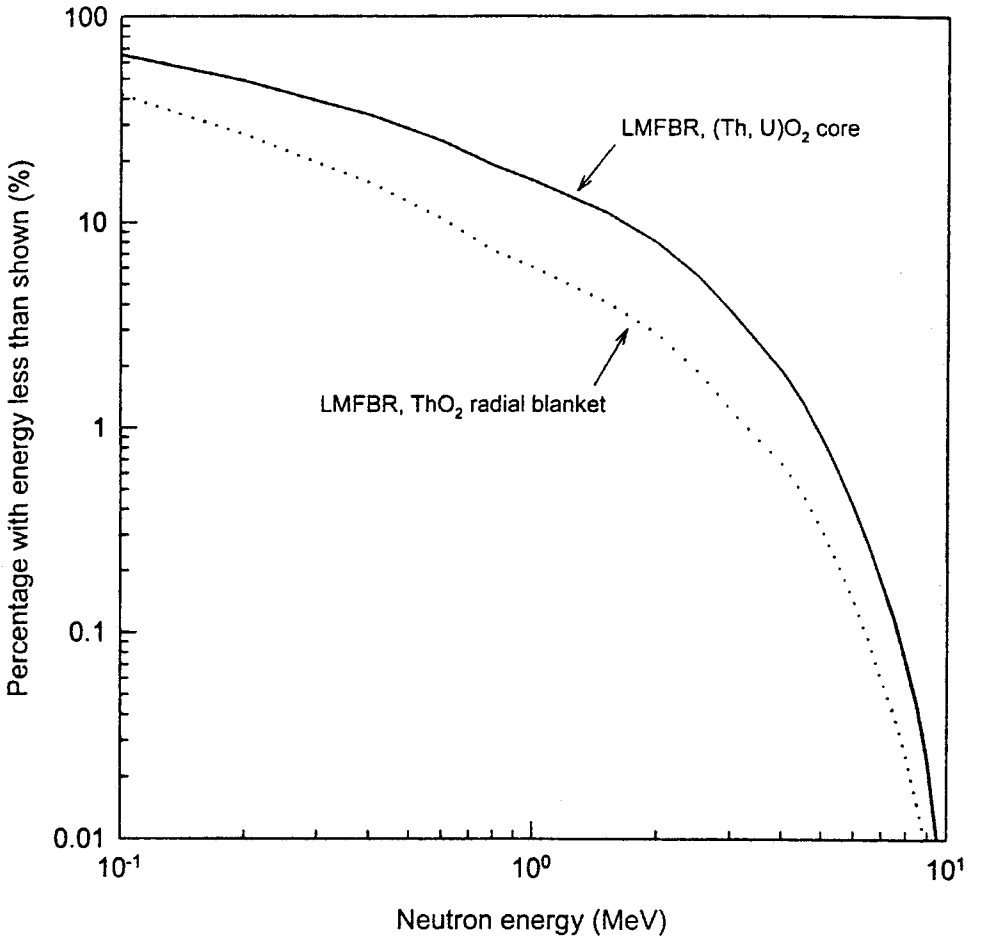

Figure 9c: Neutron-energy spectrum in LMFBR core and radial blanket.

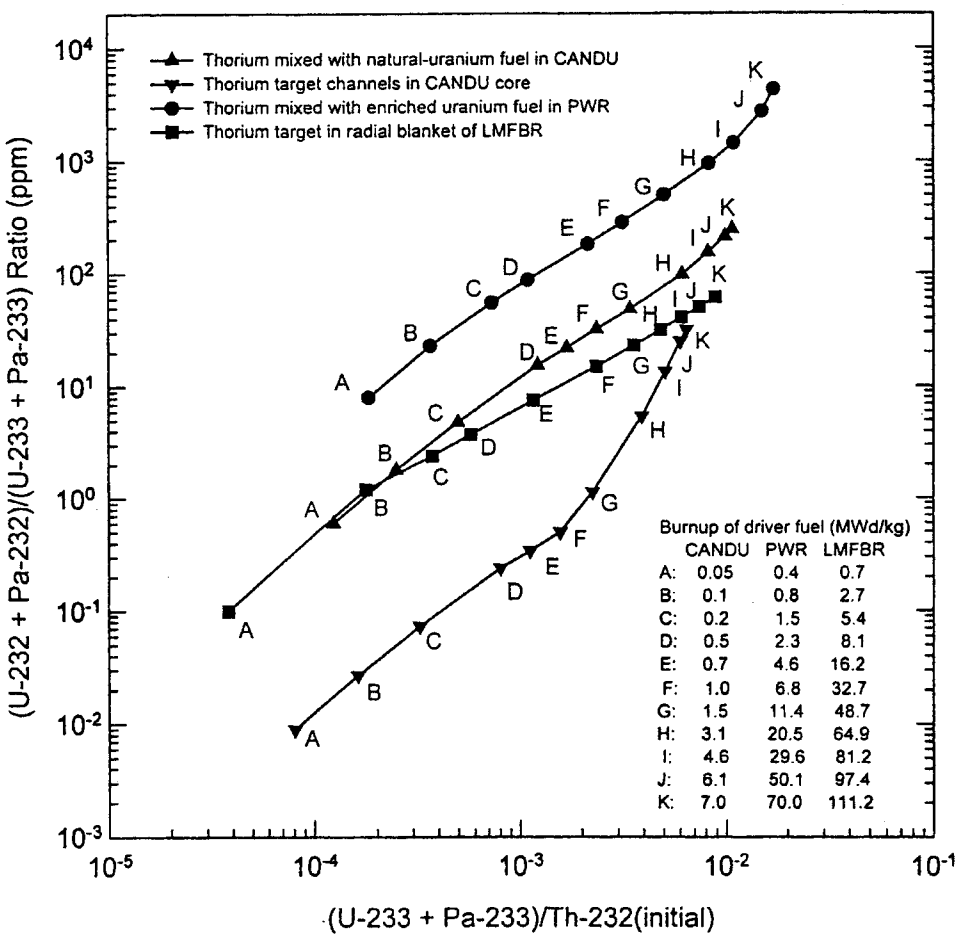

Figure 10: U-232/U-233 ratio as a function of U-233/Th-232 ratio in HWR, PWR, and LMFBR.

rium blanket compared to that in the core.

Table 3: Percentage of neutron flux in different energy intervals for fission spectrum and in HWR, PWR, and LMFBR fuel and target channels (percent).

Energy interval (in electron Volts [eV], and millions of eV [MeV]	**E ≤ 1eV**	**1 eV < E ≤ 6 MeV**	**E > 6 MeV**
Fission Spectrum	-	97.4	2.6
HWR			
Mixed Fuel(7% ThO_2)	41.0	58.0	0.4
Target channel	81.5	18.5	0.02
PWR			
Fuel (70% ThO_2, 30% UO_2)	8.4	90.8	0.8
LMFBR			
Core Fuel (ThO_2, UO_2)	-	99.6	0.4
Radial blanket (ThO_2)	-	99.9	0.1

Figure 10 shows the U-232/U-233 ratio as a function of the U-233/Th-232 concentration ratio for: U-233 production in: thorium mixed with natural-uranium fuel and in a separate thorium target channel in a HWR; a mixture of 19.5% enriched uranium and thorium in a homogenous PWR core; and in an LMFBR thorium blanket.[25]

It will be seen that, in general, U-232 contamination of the U-233 increases with burnup, reflecting the fact that two successive neutron captures are required to produce U-232. It will also be seen that contamination levels at comparable U-233/Th-232 ratios are higher for PWR's than in homogeneously fueled HWRs and lower for HWRs in which the thorium is segregated into separate “target” channels and in LMFBR blankets. To first order, these differences are explainable by differences in the presence of high-energy neutrons in the corresponding neutron spectra, as shown in Table 3.

“Clean” U-233 with a low (< 1 ppm) U-232 contamination can be produced in heavy-water reactors in mass fractions up to 0.2 percent in thorium “targets” (see Figure 10). The corresponding mass fraction in which “weapon-grade” (< 6% Pu-240 plutonium) is produced in natural uranium is 0.12 percent (see Figures 11a and 2). However, for a natural-uranium-fuel reactor, the production rate would be limited to about one quarter of that feasible for weapon-grade plutonium (see Figures 2 and 3). Plutonium containing less than 6-percent Pu-240 is considered weapon-grade, although plutonium con-

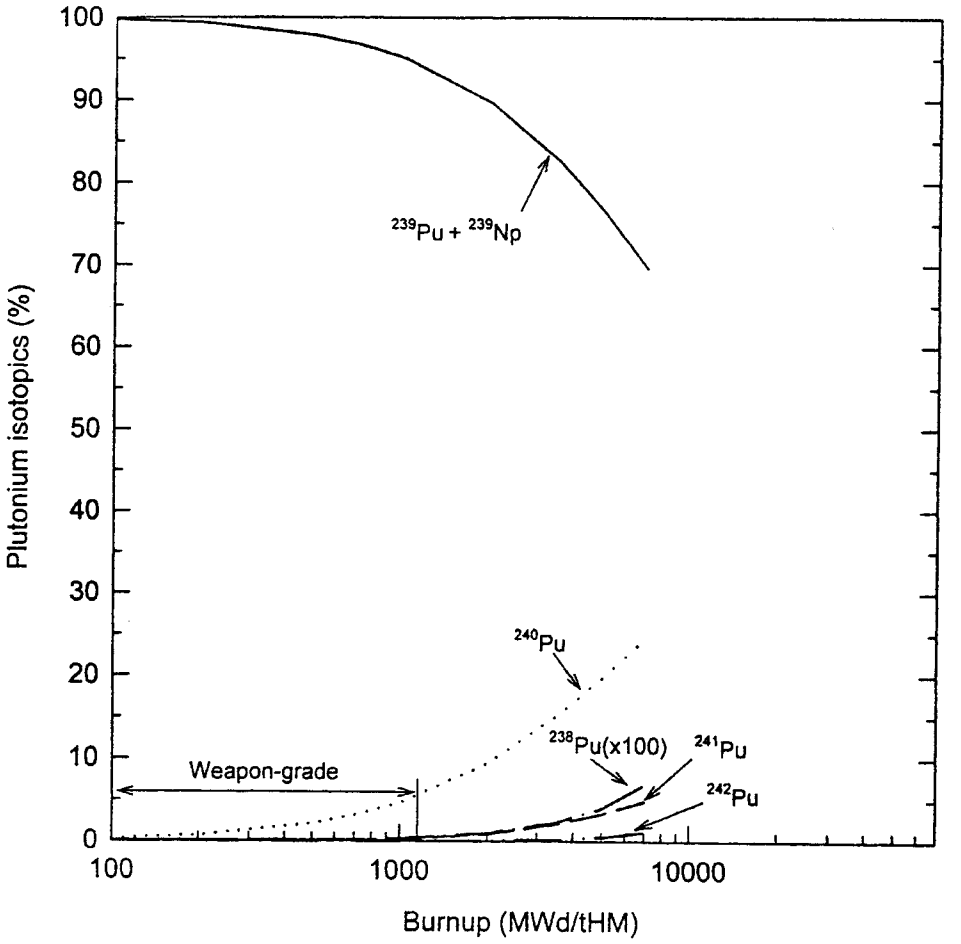

Figure 11a: Plutonium isotopics as a function of burnup in a natural-uranium fueled heavy-water reactor.

taining more Pu-240 is weapons-usable.

For LWRs with feasible target replacement schedules (on the order of ten times the frequency for maximum driver-fuel burnup) the concentration of U-232 will be above 100 ppm. At such contamination levels, remote production operations would be required to produce fuel or weapons on a large scale without incurring large occupational doses. However, it could still be feasible for a highly motivated group to make a few nuclear weapons with this material without remote processing facilities.

The U-232 contamination level in U-233 would reach about 2000 ppm in

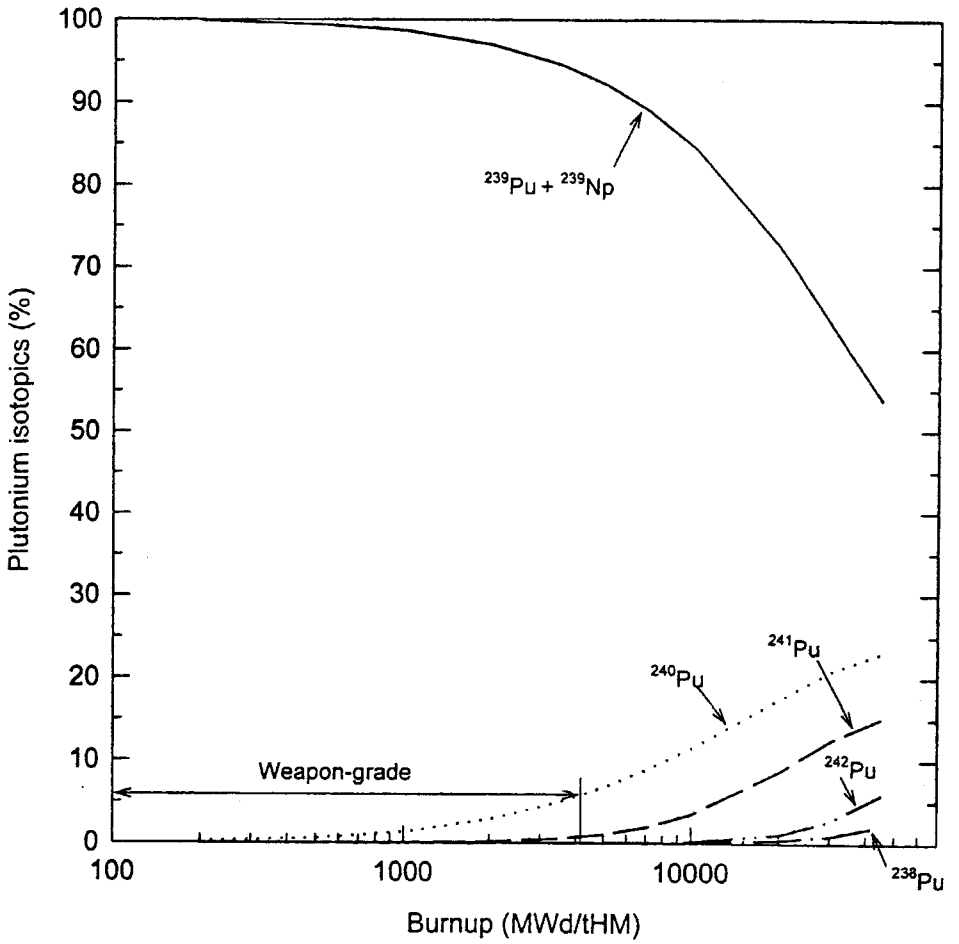

Figure 11b: Plutonium isptopics as a function of burnup in a pressurized light-water reactor (PWR) fueled with 4.5-percent enriched uranium.

LMFBR core fuel in equilibrium recycle.[26] The contamination level of the U-233 produced in LWRs fueled with mixtures of enriched uranium and thorium would be still higher but, even at several thousand ppm, the dose rate from a 5-kg sphere of U-233 would be still about an order-of-magnitude lower than that required to achieve the IAEA criterion for self-protection of 100-rem per hour at 1 meter (see Table 2).

For "fresh" U-233, i.e. U-233 cleansed of the U-232 decay product Th-228, more recently than one year, the dose rates would be proportionately smaller. Furthermore, after a U-233 "pit" for a nuclear weapon had been fabricated, it

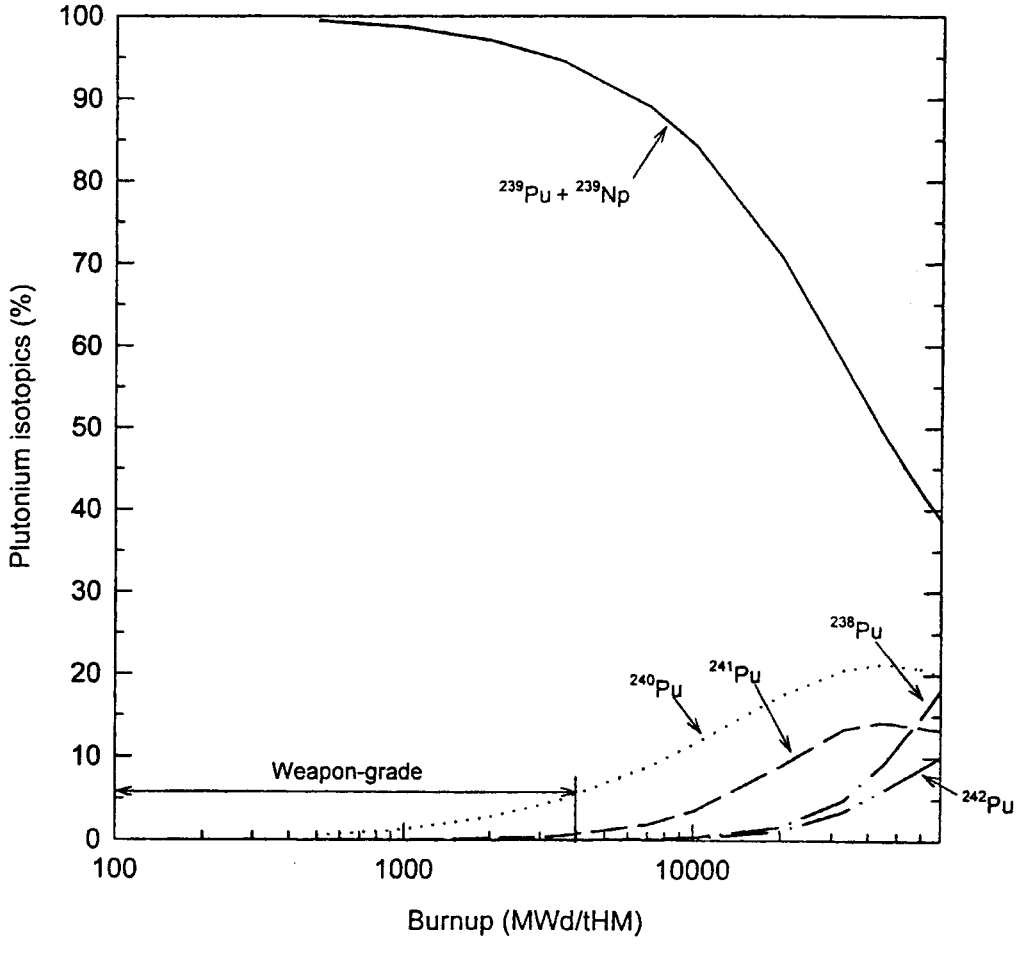

Figure 11c: Plutonium isotopics as a function of burnup in a PWR fueled with a 1:3 mixture of 19.5-percent enriched uranium and thorium.

would be practical to reduce the radiation levels to nearby personnel by an order of magnitude with a portable lead shield if the warhead design were such that the pit was solid and insertable shortly before use. Such designs were standard for safety reasons in early U.S. nuclear bombs.[27]

Conclusions

On the one hand, gamma radiation from U-232 makes the U-233 from high-burnup U-233-thorium fuel cycles more of a radiation hazard than plutonium. On the other hand, because of its low rate of spontaneous-neutron emission, U-233 can, unlike plutonium, be used in simple "gun-type" fission-weapon designs without significant danger of the yield being reduced by premature initiation of the fission chain reaction.

The necessity for remote handling of heavily U-232 contaminated U-233 in a closed fuel cycle provides a strong incentive for integration of reprocessing and fuel-fabrication. Such integration was envisioned for plutonium breeder reactors in the integral fast reactor proposal.[28] In the case of the molten-salt U-233 breeder reactor, it was proposed to have continual chemical processing of a stream of liquid fuel. Such an arrangement also offers a way to completely bypass the U-232 contamination problem because 27-day half-life Pa-233 could be separated out before it decays into U-233. [29]

In any case, no fuel cycle involving the separation and recycle of U-233 would approach the proliferation resistance of unreprocessed spent fuel from which the radiation dose rate is on the order of one thousand rem per hour at one meter for decades after discharge.[30]

Appendix A: Calculation of U-233, U232 and Plutonium Production using ORIGEN2 and MCNP

The Oak Ridge Isotope Generation and Depletion Code (ORIGEN 2.1) was used to calculate the buildup and depletion of isotopes in reactor fuel and fertile materials. ORIGEN2 is a depletion code using a matrix exponential method to calculate the production, transmutation and decay of nuclides. It is a one-group code, i.e. it comes with nuclear cross-sections which have already been convoluted with the neutron energy spectra of different reactors. A starting mix of isotopes is exposed to this neutron flux and the buildup and depletion of different species is calculated in steps of total fluence corresponding to 60-day time-steps. Many of the plots shown in this paper are obtained from correlations of isotope ratios calculated in this way.

In calculating the production rates of actinides in the fuel of natural-uranium-fueled HWRs and LEU-fueled PWRs, we used the CANDUNAU.LIB and PWRU50.LIB respectively in the ORIGEN2 cross section library. However, for production rates of actinides in thorium bearing fuels and targets in HWRs, PWRs, and LMFBRs, we modified the cross section libraries of CANDUNAU.LIB, PWRD5D35.LIB, and AMO0TTTR.LIB respectively, using set of accurate neutron fluences and one-group cross sections using the MCNP code.

MCNP is a Monte Carlo code that can calculate the transport and interaction of neutrons, photons and electrons in a three dimensional system.

ORIGEN2 was then used to do the buildup and depletion calculations for the isotopes in the fuel. The resultant ORIGEN2 output composition is then incorporated into a new MCNP input and a new set of fluxes and cross sections calculated. The entire cycle is repeated for each time-step.

Appendix B: Calculating one-group transmutation cross-sections using MCNP

MCNP. The neutron energy spectra in different core regions were calculated using simplified core models in MCNP. In this code, a cycle of calculations is initiated by introducing an initial number of neutrons into the reactor fuel with a fission-spectrum energy distribution. The code then follows the trajectory of each neutron, selecting on a random probabilistic basis in steps along the trajectory one of all the possible interactions (including none) that the neutron could have had.

Each neutron is followed until lost by absorption or leakage. Then a new set of neutrons is generated probabilistically at sources determined by reactions [fission or (n, 2n)] associated with the absorption of the first set. Thus the calculation follows the neutrons generation by generation. After a number of generations, the neutron energy and spatial distributions become insensitive to the starting distribution. Output results are then calculated by averaging the results of subsequent generations. We end the calculations when the standard deviations of the statistical fluctuation in the averages being calculated have been reduced to less than 5 percent.

The neutron multiplication factor of an assembly (k_{eff}) is calculated from the average number of neutrons produced per neutron absorbed. The production of specific isotopes per neutron absorbed in, for example, Th-232 is calculated by taking the average of the ratio of the number of atoms of that isotope produced divided by the number of neutrons absorbed in Th-232. Neutron energy spectra are calculated by adding the total path lengths of neutrons in a particular energy interval and dividing by the total neutron path-length. Neutron cross sections are calculated from the number of reactions of a specified type per unit volume (R_i) in a homogeneous region divided by the concentration of the target species (N_t) and by the total neutron path-length L_n per unit volume in that region.

$$\sigma_i = R_i/(N_t L_n)$$

HWR core models. In CANDU HWR cores the fuel is placed in "pressure tubes" carrying pressurized high-temperature heavy water coolant. A 600-MWe CANDU HWR contains 380 fuel channels (see Figure B-1[31]). The pressure tube is surrounded by an insulating, gas-filled gap and "calandria" tube. The calandria tubes are submerged in a large tank (the calandria) of cooled low-pressure heavy water moderator. Each channel contains twelve 50-cm-long fuel bundles.[32] Each fuel bundle contains 37 fuel rods, approximately 1.2 cm in diameter. Table B-1 gives the tube and fuel-element dimensions, materials and material densities.

In the MCNP calculations whose results are presented here, the fuel bundle and pressurized heavy water are treated as a homogeneous mix inside the tube and the pressure and calandria tube materials are mixed with the moderator heavy-water outside. The cross-section of the calandria is divided into square cells, as shown in Figure B-1, each of which contains one calandria tube.

In all HWR calculations, the amount of thorium added was limited by the requirement that $k_{eff} = 1.01$ at a core fuel burnup of 1000 MWd/ton-U. This is one seventh of the typical burnup in HWRs fueled with natural uranium but typical of the burnups used to produce weapon-grade plutonium from natural uranium fuel.[33]

Homogeneous-core calculations were done in single-cell approximation (see Figure B-2a). The effect of the surrounding core was simulated by imposing reflecting boundary conditions (i.e. any neutron leaving the cell was replaced by a neutron of the same energy entering at the same point from the adjoining cell with the component of its velocity vector perpendicular to the cell wall reversed). This is a reasonable approximation when the core radius is large measured in cell diameters.

For heterogeneous cores, partial-core models involving multiple cells were used. For cases where fuel and thorium target assemblies were in separate pressure tubes, we did a 3x3 "super-cell" calculation with the thorium cell in the center (see Figure B-2b). The presence of channels surrounding the super-cell was again simulated by making the outer boundaries of the super-cell neutron reflecting.

For the case with thorium channels on the periphery of the reactor, a 95-cell super-cell was used (see Figure B-2c). Here the boundaries adjoining other parts of the core were made reflecting. The boundaries that were outside of the reactor, including, the ends of the cells, were made absorbing, reflecting the neutron leakage from the core.

Table B-1: Heavy-water reactor (CANDU 600) fuel, tube and cell dimensions, materials and densities[34]

Fuel rods per bundle	37
Uranium enrichment	0.711 w/o (natural)
Fuel pellet diameter	1.217 cm
Cladding thickness	0.041 cm
Cladding and tube material	Zircalloy-4
Pressure tube inside diameter	10.363 cm
Pressure tube thickness	0.419 cm
Calandria tube inside diameter	12.878 cm
Calandria tube thickness	0.156 cm
Tube spacing (square array)	28.575 cm
Length of fuel channel	600 cm
UO_2 density	10.36 g/cm^3
ThO_2 density	9.45 g/cm^3
Zircalloy density	6.50 g/cm^3
Coolant (D_2O, 561 ^oK) density	0.81 g/cm^3
Moderator (D_2O, 346 ^oK) density	1.11 g/cm^3

Table B-2: PWR fuel pin and cell dimensions, materials and densities[35]

Fuel pellet diameter	0.819 cm
Active fuel length	365.8 cm
Uranium enrichment	4.5 w/o
Cladding thickness	0.057 cm
Cladding material	Zircalloy-4
Cell dimension	1.26 x 1.26 cm
UO_2 density	10.36 g/cm^3
ThO_2 density	9.45 g/cm^3
Zircalloy density	6.50 g/cm^3
Coolant (H_2O, 605 ^oK) density	0.64 g/cm^3

PWR and LMFBR Core Models

Since fuel pins are closely spaced with no major gaps in PWR (and LMFBR) cores, homogeneous-core calculations were done with a cell including a single fuel pin with its share of surrounding light-water (or sodium) coolant/moderator with a reflecting boundary. The specifications for the pin and cell are shown in Table B-2 (B-3).

Table B-3: 1GWe LMFBR core dimensions, materials and densities[36]

Radii	
Core	162.2 cm
Radial blanket outer edge	202.5 cm
Shield inner edge	254.6 cm
Heights	
Core	50 cm
Axial blanket	35 cm
Volume ratio (Fuel or target material/ Structure/Coolant)	
Core	40.5/22.4/37.1
Radial blanket	50.5/18.4/37.1
Material	
Fuel	$(U,Th)O_2$
Radial and axial blanket	ThO_2
Heavy metal isotopic ratio of core	
Th-232/U-232/U-233/U-234/U-235	79.31/0.04/17.22/2.98/0.45
$(U,Th)O_2$ density	9.64 g/cm^3
ThO_2 density	9.45 g/cm^3
Stainless steel (SS-304) density	0.97 g/cm^3
Coolant (Na, 823 ^oK) density	0.84 g/cm^3

Appendix C : Calculating dose rates using ORIGEN2 and MCNP

We calculated the radiation doses from 5-kg spheres of uranium and plutonium metal of varying isotopic composition (see Table C-1). Given a specified initial mix of radioisotopes, ORIGEN2 calculates, as a function of decay time, the source intensities of spontaneous neutron emissions and gamma-ray emissions – the latter grouped in 18 energy intervals.

This radiation-source data is then used as input to MCNP which performs radiation transport calculations throughout the material yielding the intensities and energy spectra of the gamma-rays and neutrons leaving the sphere. Self-shielding by the human body is simulated by assuming a 10-cm-thick water shield around the sphere. The dose rate at a point 0.5 m from the

sphere surface is then calculated using ANSI/ANS-1991 fluence-to-dose factors.[38] Statistics are accumulated until the standard-deviation uncertainty is less than 5 percent. Figure 5a and Table C-2 show the calculated buildup of the dose rates from the 5-kg spheres as a function of time.

Table C-1: Composition of 5-kg U and Pu spherical radiation sources

	Pure U-233	U-233 +1 ppm U-232	Plutonium[39]	
			Weapon-grade	Reactor-grade
Density (g/cm^3)	19.05	19.05	19.86	19.86
Sphere radius (cm)	3.97	3.97	3.92	3.92
Isotopic %				
U-232	0.0	0.0001		
U-233	100.0	99.9999		
Pu-238			0.01	1.3
Pu-239			93.80	60.3
Pu-240			5.80	24.3
Pu-241			0.35	9.1
Pu-242			0.02	5.0

Table C-2: Dose rates 0.5 m from surface of 5-kg spheres of U and Pu (mrem/hr)

Source Material	Radiation Type	0 yr	1 yr	5 yr	10 yr	15 yr
U-233 + 0 ppm U-232	Gamma	0.32	0.42	0.84	1.35	1.89
U-233 + 1 ppm U-232	Gamma	0.32	13.08	35.10	39.57	39.17
	(from Tl-208)	(0.00)	(11.12)	(29.96)	(33.48)	(32.64)
Weapon-grade Plutonium	Gamma	0.49	0.71	1.16	1.57	1.84
	Neutron	0.56	0.56	0.56	0.56	0.56
	Total	1.05	1.27	1.71	2.13	2.40
Reactor-grade Plutonium	Gamma	0.49	5.54	16.72	28.64	37.54
	(from Am-241)	(0.00)	(3.24)	(14.60)	(26.00)	(34.80)
	Neutron	2.66	2.66	2.65	2.64	2.63
	Total	3.16	8.20	19.37	31.28	40.17

NOTES AND REFERENCES

1. Calculations are done with ORIGEN2 (ORIGEN 2.1: "Isotope Generation and Depletion Code Matrix Exponential Method," [Oak Ridge National Laboratory, Radiation Safety Information Computational Center, August 1996]).

2. There are approximately 2.42 neutrons released per thermal-neutron induced fission of U-235. Of these, an average of 1.22 must be absorbed by U-235 to induce a follow-on fission, leaving 1.2 for absorption in U-238, fission products, actinides, structural, moderator and reactivity-control materials, and leakage out of the core. The relatively low light-water reactor (LWR) conversion ratio shown in Figure 2 (i.e. its delivery of a smaller fraction of excess neutrons to U-238) at low burnups reflects the facts that there is a large net flow of neutrons from fresh LWR fuel to fuel assemblies with higher burnups and the greater use of neutron absorbers in PWRs to even out swings in reactivity because they are refueled at annual or longer periods, versus continuous refueling for the HWRs. It also reflects the fact that the neutrons in HWRs are absorbed at lower energies where the fissile material capture cross sections are very high and therefore the competition from absorption on fission products is reduced (0.3 neutrons lost in the PWR vs. 0.08 in the HWR). This is partially offset by the greater absorption in HWR structural materials (0.12 neutrons vs. 0.02 in the PWR), although the absorption is less in the HWR coolant (0.03 vs 0.07 for the LWR). At higher burnups the net production of plutonium falls primarily because a significant fraction of the fissions are in plutonium.

3. Thomas Cochran et al, *U.S. Nuclear Warhead Production* (Ballinger, 1987) pp. 67-70.

4. Plutonium recovered from low-enriched pressurized-water-reactor fuel with a burnup of 33 MWd/kg and stored for 10 years before reprocessing ("Plutonium Fuel: An Assessment" (Paris: OECD/NEA, 1989), Table 9.

5. US Nuclear Regulatory Commission, "NRC Dose Limits" (http://www.nrc.gov/NRC/EDUCATE/REACTOR/09-DOSESTANDARD/index).

6. International Atomic Energy Agency, "The Physical Protection of Nuclear Material and Nuclear Facilities," INFCIRC/225/Rev.4 (http://www.iaea.org/worldatom/program/protection/index.html). Vomiting would begin within a few hours and a short-term dose of ionizing radiation could be lethal at a whole-body dose of 200 rems. Lethality within 10 days would be virtually certain above 1000 rems (*The Effects of Nuclear Weapons, 3rd edition*, Samuel Glasstone and Philip J. Dolan, eds [US Departments of Defense and Energy, 1977], Table 12.108).

7. R. Chidambaram and C. Ganguly, "Plutonium and thorium in the Indian nuclear programme," *Current Science* 70, January 10, 1996, pp. 21-35.

8. A.M. Bhagwat, K.V. Kamath, K.N. Kutty, G.R. Naik, K.K. Narayan, P.R. Pillai, G.J. Prasad, and C. Ganguly, "Radiological Safety Experience in the Fabrication of Alloy Plate Fuels Bearing 233U/Pu," *Nuclear Technology* 103 (August 1993): 246-256. Unfortunately the time since separation of the U-233 was not reported. "Person-rem" denotes the sum of the individual doses incurred by the workers.

9. A.M. Perry and A.M. Weinberg, "Thermal Breeder Reactors," *Annual Review of Nuclear and Particle Science*, 22 (1972): 317-354.

10. See. e.g., J.B. Slater, "An Overview of the Potential of the CANDU Reactor as a Thermal Breeder," (*Atomic Energy of Canada Limited*, AECL-5679, 1977).

11. See, e.g. "Final Environmental Statement, Light Water Breeder Program," (U.S. Energy Research and Development Administration, ERDA-1541, 1976).

12. See e.g. Perry and Weinberg, "Thermal Breeder Reactors."

13. R. Chidambaram and C. Ganguly, "Plutonium and thorium in the Indian nuclear programme."

14. "Thorium based fuel options for the generation of electricity: Developments in the 1990s" (Vienna, IAEA-TECDOC-1155, May 2000).

15. Manson Benedict, Thomas Pigford and Hans Wolfgang Levi, *Nuclear Chemical Engineering*, McGraw-Hill (1981): 367, 377.

16. See e.g. Alex Galperin, Paul Reichert and Alvin Radkowsky, "Thorium Fuel for Light Water Reactors – Reducing Proliferation Potential of Nuclear Power Fuel Cycle," *Science & Global Security* 6 (1997), pp. 265-290 [heterogeneous core]; J. Stephen Herring and Phillip E. MacDonald, "Characteristics of Mixed Thorium-Uranium Dioxide High Burnup Fuel" (Idaho National Engineering and Environmental Laboratory, INEEL/CON-99-00141 preprint, Nov. 13, 1998).

17. C. Rubbia et al, "Conceptual Design of a Fast Neutron Operated High Power Energy Amplifier," CERN/AT/95-44 (ET), 1995 (http://preprints.cern.ch/cgi-bin/tiff2ps?/archive/electronic/cern/preprints/at/at-95-044); C. Roche and C. Rubbia, "Some Preliminary Considerations on the Economical Issues of the Energy Amplifer," CERN/AT/95-45 (ET) (http://preprints.cern.ch/cgi-bin/setlink?base=preprint&categ=cern&id=lhc-98-012).

18. The IAEA considers plutonium containing more than 80 percent Pu-238 to be not weapons usable because of the high heat output of the short-lived (88-year half-life) Pu-238 (560 Watts/kg). However, Pu-238 is produced by neutron capture on Np-237, which is itself produced by two successive neutron captures on U-235 and then U-236. It is therefore impractical to produce Pu-238 in sufficient quantities to denature a significant fraction of the hundreds of tons of currently separated weapon- and reactor-grade plutonium.

19. Calculated using MCNP (MCNP4B2: "Monte Carlo N-Particle Transport Code System," (CCC-660 MCNP4B2, Radiation Safety Information Computational Center, January 1998).

20. For tails assay one quarter to one half of the feed assay. For a common tails assay of 0.3 percent, the ratio is about 0.3.

21. 5.7 MeV for n + U-233‡U-232 + 2n; 6.3 MeV for n + Th-232‡Th-231 + 2n.

22. ENDF/B-6.0:<http://hpngp01.kaeri.re.kr/cgi-bin/w3endf/?lab=aa&mt=Th-232%3D102&ax=auto&e0=&e1=&ay=auto&y0=&y1=&sx=&sy=>, <http://hpngp01.kaeri.re.kr/cgi-bin/w3endf/?lab=aa&mt=Th-232%3D16&ax=auto&e0=&e1=&ay=auto&y0=&y1=&sx=&sy=>.

23. ENDF/B-6.0: < http://hpngp01.kaeri.re.kr/cgi-bin/CoNquery?nuc=U233>, < http://hpngp01.kaeri.re.kr/cgi-bin/CoNquery?nuc=Th232>.

24. Examples of average neutron-flux levels in the different reactors (in units of 1014 neutrons cm-2 sec-1) are: CANDU: 2.35, PWR: 3.25, and LMFBR core ((Th,U)O2): 40.1 and radial blanket (ThO2): 5.1. (A.G. Croff and M.A. Bjerke, "Once-Through CANDU Reactor Models for the ORIGEN2 Computer Code" [Oak Ridge National Laboratory, ORNL/TM-7177, November 1980]. A.G. Croff et al., "Revised Uranium-Plutonium

Cycle PWR and BWR Models for the ORIGEN Computer Code" [Oak Ridge National Laboratory, ORNL/TM-6051, September 1978]; and A.G. Croff et al., "LMFBR Models for the ORIGEN2 Computer Code" [Oak Ridge National Laboratory, ORNL/TM-7176, October 1981].)

25. The U-232 and U-233 concentrations include respectively the U-232 precursor, Pa-232 (1.3 day half-life), and the U-233 precursor, Pa-233 (27 day half-life). Adding 1 ppm Th-230 to pure Th-232 increases the U-232/U-233 ratios compared to those without Th-230 by 0.04%, 3.5%, 3.3%, and 0.00% respectively, for thorium mixed with HWR natural-uranium fuel, a thorium target in a HWR core, thorium mixed with LEU fuel in a PWR, and thorium in the radial blanket of an LMFBR. In the case of the thorium target and blanket elements, we have assumed residence times in the reactor equal to those of the driver fuels.

26. Croff et al., "LMFBR Models for the ORIGEN2 Computer Code"

27. R.E. Kidder, "Report to Congress: Assessment of the Safety of U.S. Nuclear Weapons and Related Nuclear Test Requirments" (Lawrence Livermore National Laboratory, UCRL-LR-107454, 1991): 6.

28. See e.g. "The Design Rationale of the IFR," D. C. Wade and R. N. Hill, *Progress in Nuclear Energy*, 31, 13 (1997).

29. The designers of the molten-salt breeder reactor planned to do this so as not to lose Pa-233 to neutron capture before it decays into fissile U-233.

30. At 50 years, the dose rate is about 1000 rem/hr 1 meter from the mid-point of a spent PWR assembly and about 400 rem/hr 1 meter from the end (Dose Rate Estimates from W.R. Lloyd, M.K. Sheaffer and W.G. Sutcliffe "Irradiated Light-Water-Reactor Fuel Assemblies in Air" [Lawrence Livermore National Lab, UCRL-ID-115199, 1994]).

31. "A Study on the Direct Use of Spent PWR Fuel in CANDU Reactors: Fuel Management and Safety Analysis," (Korea Atomic Energy Research Institute, KAERI/RR-1345/93, 1994).

32. K.M. Wasywich, "Characteristics of Used CANDU Fuel Relevant to the Canadian Nuclear Fuel Waste Management Program," AECL-10463, COG-91-340, May 1993.

33. Because a CANDU can be refueled during operation, an average burnup of 1000 MWd/t-U would correspond to a discharge burnup of 2000 MWd/t-U.

34. C.E. Till and Y.I. Chang, "CANDU Physics and Fuel Cycle Analysis," (Argonne National Laboratory, RSS-TM-2, May 1977); C.A. Bollmann et al., "Environmental and Economic Performance of Direct Use of PWR Spent Fuel in CANDU Reactors," (Department of Nuclear Engineering, MIT, MIT-NFC-TR-014, June 1998).

35. J.W. Roddy et al., "Physical and Decay Characteristics of Commercial LWR Spent Fuel," (Oak Ridge National Laboratory, ORNL/TM-9591/V1, October 1985); J.S. Herring and P.E. MacDonald, "Characteristics of Mixed Thorium-Uranium Dioxide High-Burnup Fuel," June 1999 (Idaho National Engineering Laboratory, INEEL/CON-99-00141).

36. H. Matsumoto et al., "Improvement of the Prediction Accuracy of LMFBR Burnup Properties," Proceedings of International Conference on Fast Reactors and Related Fuel Cycles; FR'91, October 28 – November 1, 1991, Kyoto, Japan; A.G. Croff et al., "LMFBR Models for the ORIGEN2 Computer Code" (Oak Ridge National Laboratory, ORNL/TM-7176, October 1981).

37. H. Matsumoto et al., "Improvement of the Prediction Accuracy of LMFBR Burnup Properties," *Proceedings of International Conference on Fast Reactors and Related Fuel Cycles*; FR'91, October 28 – November 1, 1991, Kyoto, Japan.

38. "American National Standard for Neutron and Gamma-Ray Fluence-to-Dose Factors" (American Nuclear Society, ANSI/ANS-6.1.1, 1991).

39. The isotopic compositions of WGPu and RGPu are before the decay of 14-year half-life Pu241 to Am241 begins, "Management and Disposition of Excess Weapons Plutonium: Reactor-Related Options" (Academy Press, Washington, D.C., 1995), Table 2.2.