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ABSTRACT

Proliferation attributes of plutonium contained in fuel discharged from light water reactors (LWR) have been examined as they vary with burnup and cooling time. The burnup levels are 33, 50, 60, 72, and 100 GW-thermal-days per tonne initial heavy metal (GW-d/t IHM), and the cooling times are 1, 100, and 300 years after discharge. Attributes that are frequently related to proliferation risk or proliferation resistance of a fuel cycle or of spent-fuel plutonium include bare (unreflected) critical mass (BCM, kg), thermal generation (TG, W/kg or W/BCM), spontaneous neutron emission (SNS, neutrons/s per kg or per BCM), and inherent radiation barriers (RB1, cSv/hr and RB2, cSv/hr/BCM). These attributes depend on fuel burnup because of production and depletion during power operation, and they depend on cooling time because the isotopic composition of the Pu changes as isotopes decay at different rates. Plutonium in spent fuel is proliferation resistant for many years after discharge from a LWR because of its levels of TG, SNS, and RB1. However, proliferation resistance declines with the activity of the fission products contained in the spent fuel and with the decay of the Pu isotopes that produce heat and spontaneous-fission neutrons. After 300 years of cooling/decay, the spent fuel discharged from the highest burnup (100 GW-d/t IHM) studied provides a radiation barrier of only 0.024 cSv/hr (rem/hr), and a BCM of this spent-fuel Pu would produce only 5 times the thermal generation (122 W/BCM) and 16 times the spontaneous neutron generation of Weapons-grade Pu. As a result of this cooling, the proliferation resistance of a high-burnup fuel cycle diminishes after several hundred years of decay. The results presented herein are intended for use in broader, multi-attribute and multi-objective assessments of proliferation risk associated with civilian nuclear fuel cycles.

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INTRODUCTION

Recent work in several areas has included analyses of the proliferation resistance of nuclear fuel cycles, with comparisons being made on the basis of the quantities of plutonium in the spent fuel and of the qualities of that Pu from a perspective of nuclear explosives. Fuel cycles with a potential for increased proliferation resistance include high-burnup uranium-based fuel cycles,[1] uranium-thorium fuel in seed/blanket core geometries (Radkowsky Thorium Fuel, RTF),[2] and a denatured-thorium-uranium (DTU) fuel cycle coupled with accelerator-driven transmutation technology (ADTT).[3] The DTU study included a Los Alamos conceptual Accelerator Transmutation of Waste (ATW) system.[4] In addition to these recent studies, the ongoing NMMS (Nuclear Materials Management Systems) Project at LANL includes integrated assessments of alternative future nuclear energy systems.[5] These assessments require proliferation-risk metrics related primarily to the civil fuel cycle. This latter work uses a global E^3(economics-energy-environment) model that tracks temporal and regional civil-plutonium inventories,[6] that in turn are expressed in terms of a relative proliferation-risk metric based on multi-attribute utility (MAU) analyses.[7] All of these studies relate certain nuclear explosive qualities of Pu to isotopic compositions, which vary with fuel cycle, design, burnup, and cooling time. To provide a self-consistent basis for characterizing the proliferation attributes of specific reactor concepts, and to put burnup-related proliferation-risk attributes on firmer ground, we analyzed the characteristics of Pu for a range of fuel burnups and cooling times. The results reported herein pertain to a once-through LWR fuel cycle.

APPROACH

The nuclear-explosives attributes of plutonium depend on its isotopic composition, which depends on its production and depletion rate, decay history, and neutron spectrum. To analyze various grades of Pu discharged from the civil fuel cycle, a typical LWR reactor and burnup history was used. The fuel type used in this analysis applies to a Westinghouse PWR with 17x17 fuel pins per assembly and 37.5 kW/kg IHM power density, as used in the sample problem that accompanies the SAS2H depletion/shielding analysis system of the SCALE code package used to perform these computations.[8] The fuel burnup was divided into three burn cycles, each with a down time corresponding to a capacity factor of 80%. After the third burn cycle, the spent fuel was allowed to cool for 1, 100, or 300 years. The enrichment of ^{235}U in the feed fuel was appropriately varied with increasing burnup, and the isotopic makeup of the spent fuel was extracted from the SAS2H output files. That isotopic makeup was then used to compute nuclear-explosives attributes for the various Pu compositions.

NUCLEAR-EXPLOSIVES ATTRIBUTES OF PLUTONIUM

Plutonium will accumulate in cooling/storage pools in spent fuel from LWRs (currently almost 400 tn in U.S. spent fuel, more than 1,200 tn worldwide, and growing at about 60 tn/yr[9]). This plutonium is a proliferation risk concern for three reasons: 1) because of the quantity accumulating worldwide, 2) because of the forms of that Pu (in-core, in storage, separated, in un-irradiated MOX fuel, etc.); and 3) because of the "quality" of the Pu, which is measured by its usefulness for constructing nuclear explosives. Although Pu in spent fuel is not of the same isotopic quality as weapons-grade Pu, and it is initially protected by intense radiation fields (i.e., the "spent-fuel standard"), large quantities of Pu have been identified as a proliferation issue.[10] That the "spent-fuel standard" radioactivity decays with about a 100-year half-life adds to this proliferation concern.

The attributes of Pu from "advanced" fuel cycles are often compared with the attributes of different grades of Pu. The grades commonly considered are: Weapons-grade Pu (W-G Pu),[11] Reactor-grade Pu (R-G Pu),[12] and Mixed-Oxide-grade Pu (MOX-G Pu).[13] The attributes are critical mass (bare or with simple reflectors, kg Pu), thermal energy generation (W/kg), and spontaneous-neutron emission (n/s/kg); all of which affect weapon design, construction, or the probability of achieving a given nuclear-explosive yield. Additionally, the inherent radiation intensity of spent-fuel (Sv/hr at a specified distance) is used as a fourth metric to examine relative proliferation resistance. These four attributes were calculated to

examine trends and to illuminate differences between the Pu generated from high-burnup fuel cycles and W-G, R-G, and MOX-G Pu.

The values and units used in this study are unit (per unit mass) thermal-energy generation (TG, W/kg) and spontaneous neutron source (SNS, n/s/kg) strength, and the minimum mass of a given isotope of Pu required to create a bare critical assembly (Bare Critical Mass, BCM, kg Pu). The SNS, TG, and BCM metrics for pure-isotopic compositions of Pu are listed in Table I. The heat produced by a quantity of Pu is determined by the presence of α-particle-emitting isotopes (^{238}Pu, ^{240}Pu, and ^{242}Pu), while the quantity of Pu that is required to form a critical assembly is increased only by (non-fissile) ^{240}Pu and ^{242}Pu. An index of "usefulness" of a given grade of plutonium results from combining the unit TG value with the BCM to obtain an index of alpha-decay heating per critical mass (W/BCM). A second index of "usefulness" for nuclear weapons results from combining the spontaneous neutron source strength with the critical mass to obtain an index of neutron emission per bare critical mass (SNS, μBq/BCM). The third attribute of proliferation resistance relates to the radioactivity of the spent fuel and radiation doses that would be caused by that radioactivity, and is termed a radiation barrier. For this analysis, we calculated the radiation barrier (RB1) as an unshielded dose equivalent at a distance of one meter from the center of a full-length spent fuel rod. A BCM-equivalent metric is derived from RB1 by multiplying by the number of fuel rods that contain a BCM, and is designated RB2. Each of these parameters (BCM, TG, SNS, RB1, and RB2) are calculated as they vary with burnup and cooling time.

Table I. Unit Spontaneous Neutron Source (SNS), Unit Thermal Generation (TG), and Bare Critical Masses (BCM) of Plutonium Having Pure-Isotopic Compositions.

Isotope	SNS (n/s/kg)	TG (W/kg)*	BCM (kg)
Pu-238	2.60E+06	5.6E+02	10
Pu-239	2.2E+01	1.9E+00	10
Pu-240	9.1E+05	6.9E+00	36
Pu-241	4.8E+01	4.2E+00	13
Pu-242	1.7E+06	1.1E-01	92

*TG is computed using data from Ref. 14.

CALCULATION METHOD

A depletion analysis for a fuel pin for a Westinghouse PWR was used as a Baseline for these studies. The SAS2[15] driver from SCALE 4.[8] was used for a 17x17-pin PWR fuel assembly with borated water to compute criticality, burnup, fuel depletion, and actinide production, as follows: a) cross sections are computed by BONAMI[16] and NITAWL;[17] b) XSDRN-PM[18] computes criticality and neutron spectra for a one-dimensional assembly; and c) the neutron spectra are then used to collapse the cross sections to a single neutron energy group for a depletion and decay analysis with ORIGEN-S.[19] Fuel with 3.2%-enriched UO_2 exposed to a burnup of 33 GW-thermal-days per kg (GW-d/t IHM) and cooled for one year served as the Baseline. In addition, because of interest in long-term interim or retrievable storage for used fuel, the same spent fuel was cooled for 100 and 300 years. For fuel with greater burnup, similar computations were made with the enrichment changed to achieve the higher burnup. The enrichment was taken as proportional to the burnup, and the following higher burnups were examined: 50 GW-d/t IHM (5% enriched), 60 GW-d/t IHM (6% enriched), 72 GW-d/t IHM (7.2% enriched), and 100 GW-d/t IHM (10% enriched). The isotopic compositions of spent-fuel Pu from these analyses were then used to compute bare critical masses with the KENO-Va Monte Carlo neutronics code.[20]

Both TG and SNS for plutonium of a given isotopic mixture were taken as mass-fraction-weighted averages. The radiation protection attributes of spent fuel were calculated by the product of energy-dependent, γ-ray production rates (per fuel rod) with KERMA energy deposition values[21] to produce a

dose equivalent rate (RB1, cSv/hr or rem/hr per fuel rod). That dose rate is then multiplied by a linear-source geometric factor[22] to produce the dose equivalent rate per fuel rod at a distance of one meter from the mid-point of the fuel rod. To estimate the impact of processing sufficient fuel to separate a BCM of Pu, the dose rate per fuel rod is divided by the mass of Pu per fuel rod, then multiplied by the mass of a BCM to produce a dose rate per BCM (RB2, cSv/hr/BCM or rem/hr/BCM).

RESULTS

Baseline Compared to Other Grades of Plutonium

The isotopic composition of Pu for the Baseline (e.g., 3.2%-enriched, 17x17-pin assembly, Westinghouse PWR, 33 GW-d/t IHM) is given in Table II, along with compositions of other grades of Pu. These compositions were used to compute parameters for comparison of proliferation attributes of the different grades of plutonium. The values of BCM, TG, and SNS are listed in Table II for the Baseline, W-G, R-G, and MOX-G Pu. For comparison with an advanced fuel cycle, the computed values for the Pu from seed-fuel elements of the Radkowsky Thorium Fuel (RTF)[2] is also included.

Table II. Isotopic Compositions (Weight Fractions) and Proliferation Attributes of Five Grades of Plutonium.

	Plutonium grade				
	W-G (Ref. 23)	R-G* (spent fuel) (Ref. 24)	MOX-G (feed) (Ref. 13)	RTF seed (spent fuel) (Ref. 2)	Baseline PWR (spent fuel)
Burnup (GW-d/t IHM)		33	33	180	33
Pu-238 fraction	0.00012	0.024	0.019	0.065	0.016
Pu-239 fraction	0.938	0.584	0.404	0.465	0.565
Pu-240 fraction	0.058	0.240	0.321	0.225	0.238
Pu-241 fraction	0.0035	0.112	0.178	0.155	0.128
Pu-242 fraction	0.00022	0.039	0.078	0.090	0.054
BCM (kg)	10.7	13.5	15.8	14.4	13.7
TG (W/kg)	2.3	16.6	14.3	39.3	12.1
SNS (μBq/kg)	0.06	0.90	0.91	2.02	0.71
TG (W/BCM)	24.1	224.1	226.5	564.8	166
SNS (μBq/BCM)	0.60	12.1	14.4	29.1	9.8

*3.2% enriched fuel, 150 days cooling, different core and spectrum than Baseline.

Proliferation Attributes versus Burnup for LWR Fuel Cooled for One Year

Compositions of Pu and the proliferation-risk indices BCM, TG, and SNS are listed in Table III for spent fuel of a range of burnups for a one-year cooling period. Although the BCM changes by only 3% over the burnup range examined, the TG and SNS increase by 295% and 237%, respectively. This thermal generation in a mass of Pu indicates how much heat would be generated by comparable nuclear explosives. The TG for the short cooling times for high-burnup fuel, 646 W/BCM, is about 27 times the TG in a critical mass of W-G Pu, and also exceeds the heating in Pu from LWRs, that in MOX, and that in RTF spent-seed fuel. This is sufficient heat to cause thermal management problems: any attempt to build a weapon from this material would require engineering solutions to remove decay heat to prevent degradation of weapon performance. The impact of this thermal generation on the quantification of proliferation risk will be examined in future studies under the NMMS project at LANL.[5]

Table III. Isotopic Compositions and Proliferaion Attributes of Pu from Spent Fuel Cooled for One Year (for High-Burnup Fuels, Enrichment of Fresh Fuel is Proportional to Final Burnup).

	Burnup (GW-d/t IHM)				
	33	50	60	72	100
Pu-238 fraction	0.016	0.029	0.038	0.050	0.078
Pu-239 fraction	0.565	0.533	0.518	0.502	0.470
Pu-240 fraction	0.238	0.233	0.230	0.226	0.217
Pu-241 fraction	0.128	0.139	0.142	0.145	0.147
Pu-242 fraction	0.054	0.066	0.072	0.078	0.088
TG (W/kg)	12.1	19.3	24.4	30.7	46.5
SNS (μBq/kg)	0.71	1.07	1.31	1.61	2.35
BCM (kg)	13.7	13.9	14.0	14.0	14.1
RB1 (cSv/hr)	89	99	104	109	117
TG (W/BCM)	166	269	342	432	656
SNS (μBq/BCM)	9.8	14.8	18.3	22.6	33.1
RB2 (Sv/hr/BCM)	731	687	670	646	600

Proliferation Attributes versus Burnup for LWR Fuel Cooled for 100 and 300 Years

Similar isotopic compositions and proliferation-risk attributes were computed for Pu in spent fuel that has decayed for 100 years or 300 years, as it would exist in a repository or monitored retrievable storage facility. The isotopic compositions and the proliferation-risk attributes are listed in Table IV for 100-year spent fuel and in Table V for 300-year spent fuel.

Table IV. Isotopic Compositions and Proliferaion Attributes of Pu from Spent Fuel Cooled for One-Hundred Years.

	Burnup (GW-d/t IHM)				
	33	50	60	72	100
Pu-238 fraction	0.009	0.016	0.021	0.027	0.044
Pu-239 fraction	0.653	0.626	0.613	0.598	0.567
Pu-240 fraction	0.275	0.279	0.279	0.280	0.281
Pu-241 fraction	0.001	0.001	0.001	0.001	0.001
Pu-242 fraction	0.062	0.078	0.086	0.093	0.107
TG (W/kg)	7.9	12.0	14.8	18.4	27.4
SNS (μBq/kg)	0.58	0.79	0.94	1.12	1.56
BCM (kg)	13.9	14.4	14.4	14.6	14.8
RB1 (cSv/hr)	0.019	0.023	0.025	0.028	0.034
TG (W/BCM)	110	172	213	268	405
SNS (μBq/BCM)	8.0	11.4	13.5	16.3	23.1
RB2 (cSv/hr/BCM)	17.9	19.3	20.0	20.8	22.3

Table V. Isotopic Compositions and Proliferaion Attributes of Pu from Spent Fuel Cooled for Three-Hundred Years.

	Burnup (GW-d/t IHM)				
	33	50	60	72	100
Pu-238 fraction	0.002	0.003	0.004	0.006	0.009
Pu-239 fraction	0.660	0.639	0.627	0.614	0.592
Pu-240 fraction	0.275	0.278	0.281	0.283	0.287
Pu-241 fraction	0.000	0.000	0.000	0.000	0.000
Pu-242 fraction	0.063	0.080	0.088	0.097	0.112
TG (W/kg)	4.16	5.02	5.62	6.40	8.38
SNS (μBq/kg)	0.404	0.475	0.519	0.572	0.694
BCM (kg)	14.0	14.3	14.5	14.7	15.1
RB1 (cSv/hr)	0.012	0.015	0.017	0.019	0.024
TG (W/BCM)	58	72	82	94	127
SNS (μBq/BCM)	5.7	6.8	7.5	8.4	10.5
RB2 (cSv/hr/BCM)	11.5	12.9	13.6	14.6	16.6

Impact of Cooling Time on Proliferation Attributes of Pu

The effect of longer cooling time on the three proliferation metrics, TG, BCM, and SNS, is illustrated in Figures 1, 2, and 3, which show the variations with burnup. The BCM parameter (Figure 1), varies less than 10% with burnup or cooling, and is generally about 30-40% greater than the BCM of W-G Pu. The BCM at 300 years is 10% greater than at 1 year for the highest-burnup Pu, whereas it increases only 3% after 300 years for the Baseline (33 GW-d/t IHM) case.

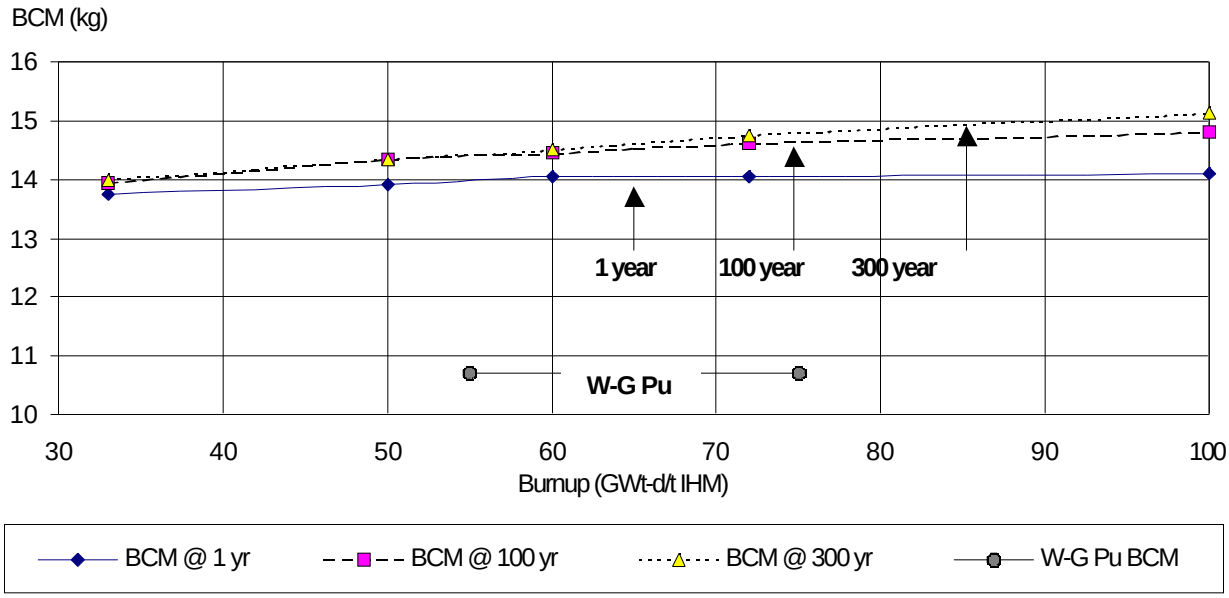

Figure 1. Bare Critical Mass (BCM) of spent-fuel Pu versus burnup and decay time.

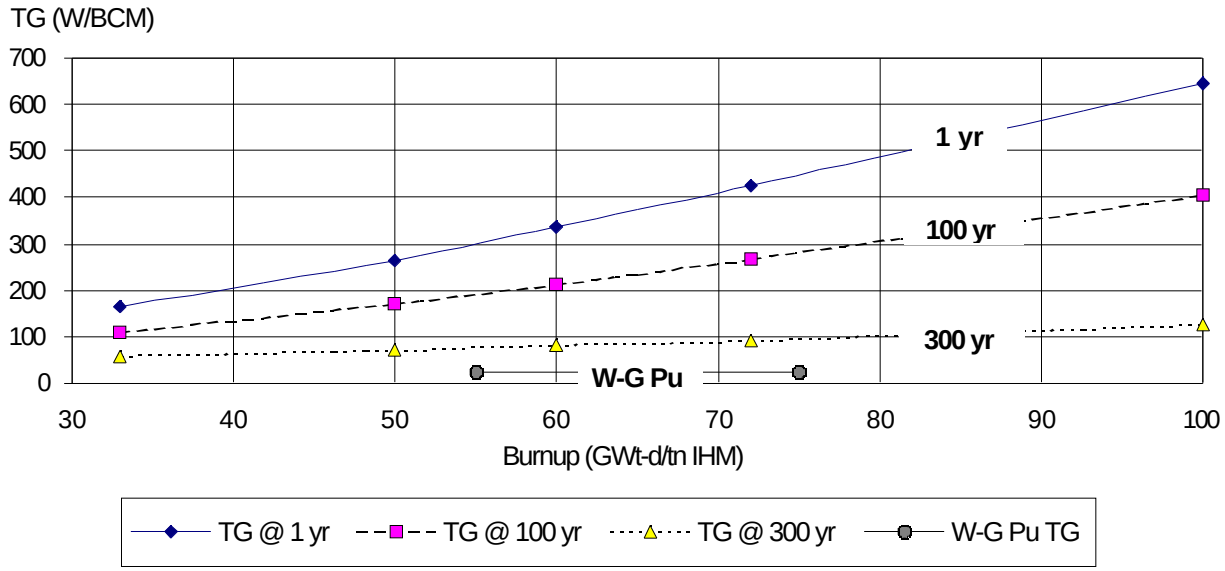

Figure 2. Thermal Generation (TG) of spent-fuel Pu versus burnup and decay time.

The relative increase in TG with burnup and the relative decrease with cooling time are illustrated in Figure 2. Compared to the Baseline case after one year of cooling, TG increases by a factor of about four over the range of burnups studied and decreases by about one-third over the range of cooling times, even though the BCM varies by only 3-10% (Figure 1). The TG of the highest-burnup fuel (100 GW-d/t IHM) decreases by a factor of five with cooling time, so that the cooled Pu produces just 125 W/BCM after the spent fuel has cooled for 300 years. Thus, the cooled high-burnup Pu produces less heat than the Baseline spent fuel at one year (166 W/BCM). Thermal energy in this 300-year-old high-burnup grade of Pu probably would not produce a substantial deterrent for nuclear-weapon manufacturing. The TG values for Pu from high-burnup LWR spent fuel are high compared with W-G Pu, but after long decay times, even these values are not high compared to fresh MOX fuel. Handling this Pu and fabricating a weapon from it would not be prevented by thermal generation. Note also that Pu from low-burnup spent fuel that has cooled for 300 years produces on the order of the same thermal energy as weapons-grade Pu. This is also a characteristic of very-low-burnup fuel that is discharged from the first unloading of a new reactor, so that its its proliferation resistance is reduced.

In contrast to the one-third to one-fifth decrease in TG, the SNS decreases less with cooling time (Figure 3), and for spent-fuel Pu that has cooled for 300 years, the SNS is still substantially greater than that of W-G Pu (11 times for 50 GW-d/t IHM). Even after hundreds of years of cooling, the SNS remains a substantial source of neutrons that could pre-initiate a chain reaction during the assembly of a mass of Pu, which could reduce its yield significantly.[25] Because of this increased SNS, the probability of a nuclear explosive producing a low yield (less than 1 kiloton) is estimated to increase from 6% for W-G to 17% for 300-year Baseline case Pu, to 30% for the highest-burnup, longest-cooled Pu. In addition, the probability of producing the full design yield is only about 12% for the one-year Baseline Pu compared to 88% for W-G Pu. For Pu extracted from the highest-burnup spent fuel after one year of cooling, the low-yield probability is 67% and the chances of obtaining the design yield are less 0.1%.

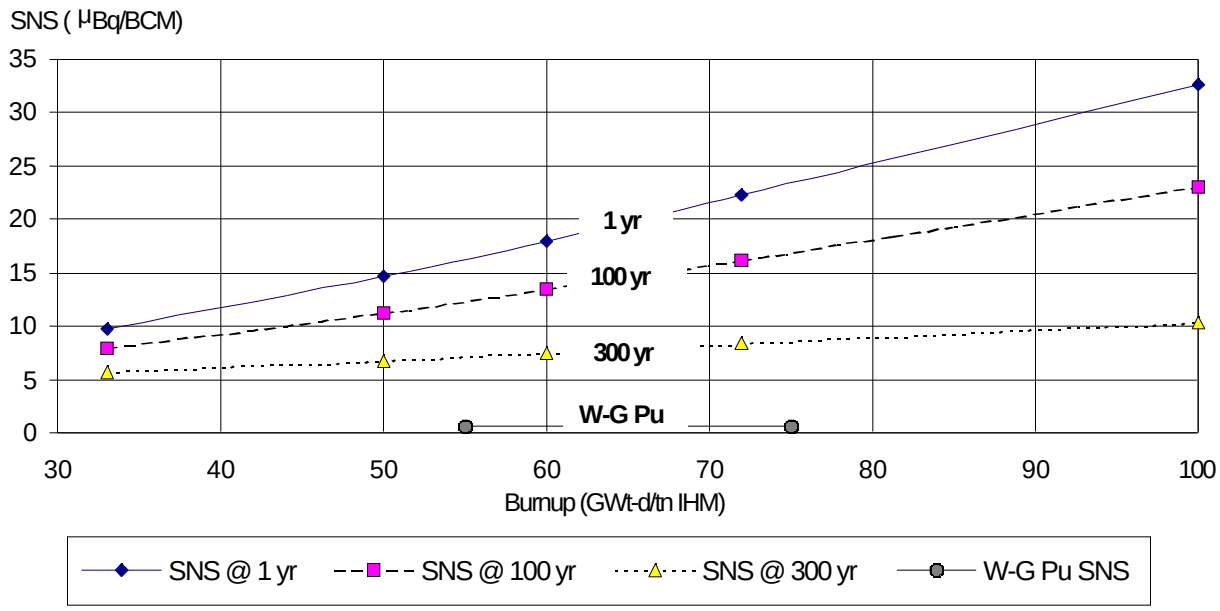

Figure 3. Spontaneous Neutron Source (SNS) of spent-fuel Pu versus burnup and decay time.

Radiation Protection versus Burnup of LWR Fuel

The emission of γ-ray radiation from used nuclear fuel decreases its attractiveness for use in obtaining nuclear explosives material, at least when the fuel is recently discharged. The radiation dose at a distance of 1 meter produced by an exposed (17x17) assembly of fuel rods would be deadly within minutes. However, after 300 years of decay the radiation dose rate is reduced by a factor of about 10,000, to the point that simple radiation protection measures could eliminate hazards associated with its handling. Radiation barrier values (RB1) versus burnup are shown in Figure 4 for spent fuel that has decayed for 1, 100, and 300 years. The RB1 value increases from the lowest to the highest burnup (30% at one year and 100% at 300 years), but decreases almost four orders of magnitude with long decay times. For 50 GW-d/t IHM spent fuel, the radiation barrier at one meter from the mid-point of a single, unshielded fuel rod (RB1) is on the order of 100 cSv/hr (rem/hr) 1 year after discharge, but that exposure declines to 0.023 cSv/hr after 100 years and 0.015 cSv/hr after 300 years.

However, the radiation barrier from the fuel required to separate a BCM (RB2) decreases with burnup for short cooling times, because the increased radiation emission is accompanied by a greater increase in Pu production per unit of spent fuel. Because of the higher initial ^{235}U concentration in high-burnup fuel, less of the Pu is burned early in the fuel cycle. Although the radiation barrier for a single fuel rod appears to be manageable, the radiation barrier produced by sufficient fuel rods to separate 1 BCM is about 1,000 Sv/hr, which would be lethal with an exposure of less than a minute. Again, this radiation barrier decreases almost four orders of magnitude for long decay times, so that even an hour of exposure to an unshielded spent fuel assembly that has cooled for 300 years would produce no detectable immediate radiation effects.

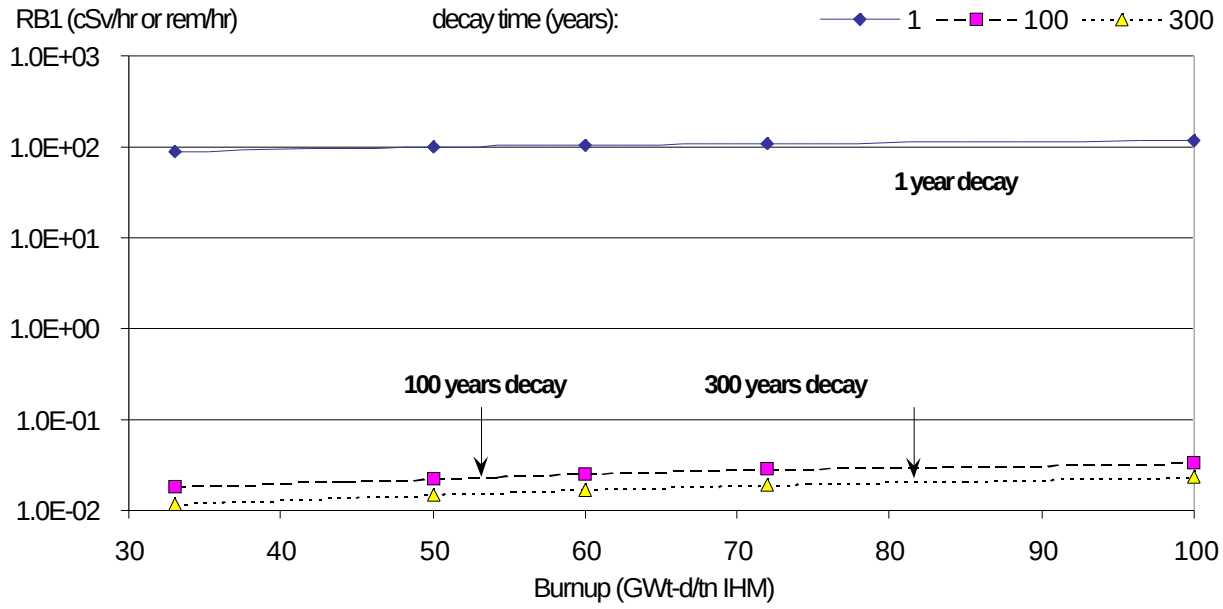

Figure 4. Radiation Barrier (RB1, cSv/hr) versus burnup and decay time for LWR spent-fuel plutonium at 1 meter from the mid-point of a single, unshielded fuel rod.

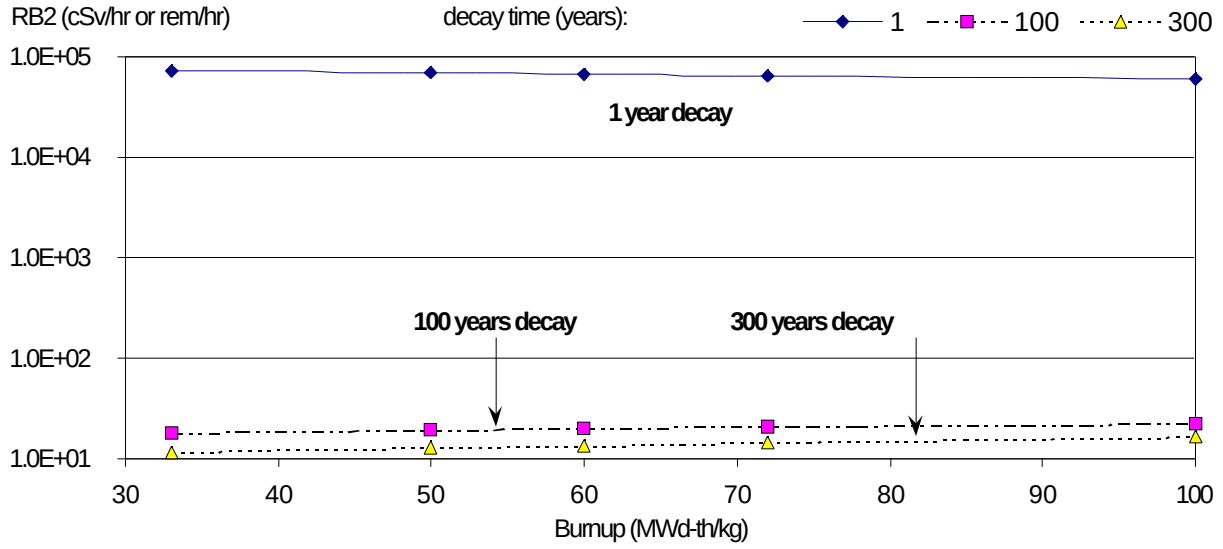

Figure 5. Radiation Barrier (RB2, cSv/hr/BCM) versus burnup and decay time for LWR spent-fuel plutonium that contains one BCM.

SUMMARY

The attributes of plutonium that is contained in used fuel discharged from light water reactors (LWR) has been examined to provide a technical basis for future quantitative assessments of proliferation risk from the civil fuel cycle. These attributes are compared for several levels of burnup and for varying periods of radioactive decay or cooling after discharge. The burnup levels are 33, 50, 60, 72, and 100 GW-d/t IHM, and the cooling times are 1, 100, and 300 years after discharge. Attributes that are frequently related to proliferation risk or proliferation resistance of a fuel cycle or of spent-fuel plutonium

include critical mass (BCM, kg), thermal generation (TG, W/kg or W/BCM), spontaneous neutron emission (SNS, μBq/kg or μBq/BCM), and inherent radiation barriers. To estimate the proliferation resistance posed by radiation barriers, we calculated the dose equivalent one meter from the mid-point of a single, unshielded fuel rod (RB1, cSv/hr) and the dose equivalent produced by enough fuel rods to separate one BCM (RB2, cSv/hr/BCM). These attributes depend on fuel burnup because of production and depletion during power operation, and they depend on cooling time because the isotopic makeup of the Pu changes because of different decay rates of the isotopes. Plutonium in spent fuel is proliferation resistant for many years after discharge from a LWR because of its levels of TG, SNS, RB1 and RB2. For 50 GW-d/t IHM spent fuel that has cooled for one year, the values of these proliferation-risk indices are: BCM–13.9 kg, TG–269 W/BCM, SNS–14.8 μBq/BCM, RB1– 99 cSv/hr, and RB2–687 cSv/hr/BCM. However, proliferation resistance declines with the activity of the fission products contained in the spent fuel and with the decay of the Pu isotopes that produce heat and spontaneous neutrons. After 300 years of cooling/decay, the spent fuel discharged from the highest burnup studied (100 GW-d/t IHM) provides a radiation barrier of only 0.024 cSv/hr (rem/hr) for a single fuel rod, and a BCM of this spent-fuel Pu would produce only 5 times the thermal generation (122 W) and 16 times the spontaneous neutron generation of weapons-grade Pu. In addition, all the fuel rods required to separate a BCM would produce a combined dose rate of only 16 cSv/hr (16 rem/hr). As a result of this cooling, the proliferation resistance of even a high-burnup fuel cycle is appreciably diminished after several hundred years of decay.

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