

Impact of High Burnup on PWR Spent Fuel Characteristics

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Abstract—Reducing the burden of management of spent nuclear fuel is important to the future of nuclear energy. The impact of higher pressurized water reactor (PWR) fuel burnup is examined in this paper from the perspective of its impact on spent-fuel radioactivity, decay heat, and plutonium content. The necessary fresh fuel enrichments to achieve high burnup in PWRs with the same three-batch operation scheme are first computed; then, characteristics of the spent fuel are determined. The increase in decay heat with burnup is found to be generally less than linear. Although each high-burnup fuel assembly would be hotter and more radioactive, the total decay heat to be removed or accommodated in storage is less for the same electricity production. If the time window before 150 yr after discharge can be excluded from impacting a repository, significant savings in its capacity can be realized with high-burnup fuel. The high-burnup fuel is more proliferation resistant because of reduced total plutonium production per kilowatt hour and because of higher content of less desirable plutonium isotopes, such as ^{238}Pu . The fuel cycle cost can be slightly reduced by increasing burnup until it reaches a shallow minimum near 70 MWd/kg. Higher burnups would require one-time changes to the limits on enrichments that can be handled in most commercial fuel fabrication facilities. Changing the waste fee to base it on the amount of radioactivity in the spent fuel would enhance the economic benefit of high burnup.

I. INTRODUCTION

Highly radioactive for many thousands of years after its use in nuclear power reactors, spent nuclear fuel has been a major public concern that clouds the potential for future expansion of nuclear energy. Although it is commonly recognized by the scientific and engineering communities that current technology should be more than adequate for isolating radioactive wastes from the biosphere and for protecting public health and safety, the spent-fuel concern remains an active societal and political issue. There are two main worries about the spent fuel: (a) the management of the long-lived, highly radioactive materials including monitoring, storage, handling, transportation, and disposition and (b) nuclear security and proliferation potential. The average annual discharge of spent fuel in the United States is ~2000 tonnes of heavy metal (about 7000 assemblies). By 2030, accu-

mulation of spent fuel will reach 80 000 tonnes of heavy metal (about 293 000 assemblies).¹ The U.S. government is responsible for the final disposal of the spent nuclear fuel according to the Nuclear Waste Policy Act. Commercial nuclear utilities have no obligation for ultimate spent-fuel disposal except for paying 1 mill/kW·h of electricity generated after April 7, 1983, into a special fund controlled by the U.S. Congress. However, storage of spent fuel prior to waste disposal is still the responsibility of the individual utility. The opening of the first U.S. geologic repository at Yucca Mountain, Nevada, is now expected to be in 2010. On the other hand, nuclear security and proliferation concerns are becoming more important since the September 11, 2001, incident. While the terrorists can choose less protected targets, in theory the highly radioactive, existing nuclear spent-fuel assemblies, which are widely spread across the world, could be potential terrorist targets. A “dirty bomb” can be made by packing radioactive materials from the spent nuclear fuel with conventional explosives if a terrorist group is

well equipped for remote handling. The plutonium in the spent fuel can also be chemically separated and diverted as weapons materials.² Increasing the difficulty of conducting the chemical separation in the radioactive environment and also producing less plutonium are possible via the higher burnup in pressurized water reactors (PWRs).

There are several approaches to reducing the rate of spent-fuel production,¹ among which a high-burnup strategy is a realistic and cost-effective one. In fact, the trend of pursuing higher-discharge fuel burnup has persisted ever since the first large-scale commercial PWR of Yankee Rowe in 1960. Nowadays, typical PWR discharged fuel burnup is ~ 50 MWd/kg, whereas in the 1970s and 1980s, the fuel burnup was ~ 33 MWd/kg. However, it should be recognized that this historical trend of increasing fuel burnup is motivated by economics, not spent-fuel management. The higher fuel burnup allows the reactor to operate for a longer time between refueling outages, thus improving the plant capacity factor. The trend of burnup increase is likely to be incremental in the future because of smaller economic incentives. The current attention in the nuclear industry is focused on power uprates, which might encourage a modest burnup increase. However, the government is obviously more concerned about spent fuel. The waste disposal repository design has to incorporate the appropriate spent-fuel characteristics as initial conditions. Although it is clear that high burnup is beneficial in terms of reducing the volume and mass of spent fuel per unit of electricity generation, the corresponding repository storage volume savings is still uncertain because of the higher decay heat generation and radioactivity associated with each assembly of the higher-burnup spent fuel.

In this paper, several representative PWR burnup levels for UO₂ fuel (including past, present, and future projected values) are chosen, based on which the spent-fuel characteristics are examined, including isotopics, radioactivity, decay heat, and radiotoxicity. Note that all spent fuel characteristics reported are for the fuel only without considering most effects related to the repository environment. System modeling of the spent fuel in the repository requires consideration of possible nuclide diffusion into the environment as well as long-term physical and chemical evolution. The work in this paper served as a technical basis for the nuclear waste discussion of a recently released Massachusetts Institute of Technology report on the future of nuclear power.³

II. CALCULATION PROCEDURES AND COMPUTER CODES

Discharged fuel burnup depends on the core management and the reload fresh fuel enrichment. Typical four-loop Westinghouse PWR cores employ a three-

batch fuel management scheme. It is assumed in this paper that the three-batch core management scheme is kept the same for all burnup levels. The 18-month fuel cycle is employed for the current burnup level of 50 MWd/kg only. The functional dependence of the discharged burnup on the average reload enrichment is calculated using the Studsvik core management system code package including CASMO-4, TABLES-3, and SIMULATE-3 (Refs. 4, 5, and 6, respectively). The licensing-level, commercial Studsvik code package is capable of performing steady-state light water reactor (LWR) core reload analysis and is being widely used by nuclear utilities to do routine fuel management calculations. CASMO-4 is a multigroup, two-dimensional transport, lattice physics code that generates two-group cross sections and other parameters (such as discontinuity factors) for each type of fuel assembly. TABLES-3 then reads CASMO-4 output card image files and generates a binary SIMULATE-readable library. As a two-group, three-dimensional nodal code, SIMULATE-3 can perform core reload calculations based on the generated library.

Table I shows the six cases analyzed for PWR spent fuel. The discharge burnup increase is achieved via a higher reload enrichment. Note that the current commercial enrichment licensing limit for most commercial fuel fabricators is 5 wt%. Hence, this barrier needs to be broken before the next higher-burnup level (70 MWd/kg) can be achieved. Although it might be expensive to upgrade the existing fuel fabrication facilities to allow higher enrichment, the obstacle is not deemed as insurmountable because the upgrade is a one-time change and can be absorbed as long as long-term requirements persist.

Spent-fuel isotopic depletion and decay analysis is subsequently performed by MCODE-1.0 (Ref. 7) and ORIGEN2 (Ref. 8), and typical Westinghouse PWR parameters, such as the moderator temperature and the power density, are assumed. MCODE-1.0 is a MCNP/ORIGEN coupling program, which uses the continuous-energy, Monte Carlo MCNP-4C code⁹ to provide the neutron flux and one-group cross sections for ORIGEN2 depletion calculations. Developed at the Oak Ridge National Laboratory, ORIGEN2 is a one-group, point depletion and radioactive decay code. The nuclide representation

TABLE I
High-Burnup Spent-Fuel Cases

Case Number	Burnup (MWd/kg)	Enrichment (wt%)	Description
1	33	3.1	Early-days discharge burnup
2	50	4.5	Contemporary burnup level
3	70	6.3	Foreseeable future
4	100	9.1	Future burnup level
5	120	11.2	Far-future burnup
6	150	14.3	Remote future

in ORIGEN2 consists of about 1700 nuclides. Thus, this code is ideal for calculating spent-fuel characteristics. Accurate burnup calculations are also achieved with the help of the advanced physics code MCNP, which routinely updates the one-group cross sections of the most important 130 fission products and 23 actinides in the fuel (accounting for >99.5% of total neutron absorptions through the entire burnup range). The detailed fuel power history including any refueling outage is ignored; constant power over the fuel depletion is assumed. Actually, given the same discharge burnup, the spent fuel has only ~10 yr of memory after discharge about the fuel operating history, after which the spent-fuel characteristics become close enough to be indistinguishable.¹⁰

III. SPENT-FUEL STORAGE AND DISPOSAL

III.A. Introduction

Spent-fuel radioactivity consists of three components:

1. actinides and their decay daughters
2. fission products from splitting of heavy metal atoms

3. activation products including activated structural materials.

Among the three groups of nuclides, the activation products have much smaller contributions to the total decay heat and radiotoxicity. Therefore, they are ignored in the following discussions. However, because of the other two categories, by the U.S. Nuclear Regulatory Commission definition, spent nuclear fuel belongs to the category of high-level waste.

Generally speaking, the actinides have very long half-lives, on the order of tens of thousands of years, and long decay chains. In contrast, most fission products have short half-lives (tens of years) and short decay chains. Therefore, the short-term spent-fuel characteristics are governed by fission products while the long-term behavior is governed by actinides. Combined together, the spent nuclear fuel remains highly radioactive for many thousands of years. Figures 1 and 2 show the radioactivity as a function of time after discharge for two specific types of spent fuel at 33 and 50 MWd/kg (past and present burnup levels in Table I). The 33 MWd/kg spent fuel dominates existing PWR spent-fuel inventory because of historical accumulation, and the 50 MWd/kg fuel is

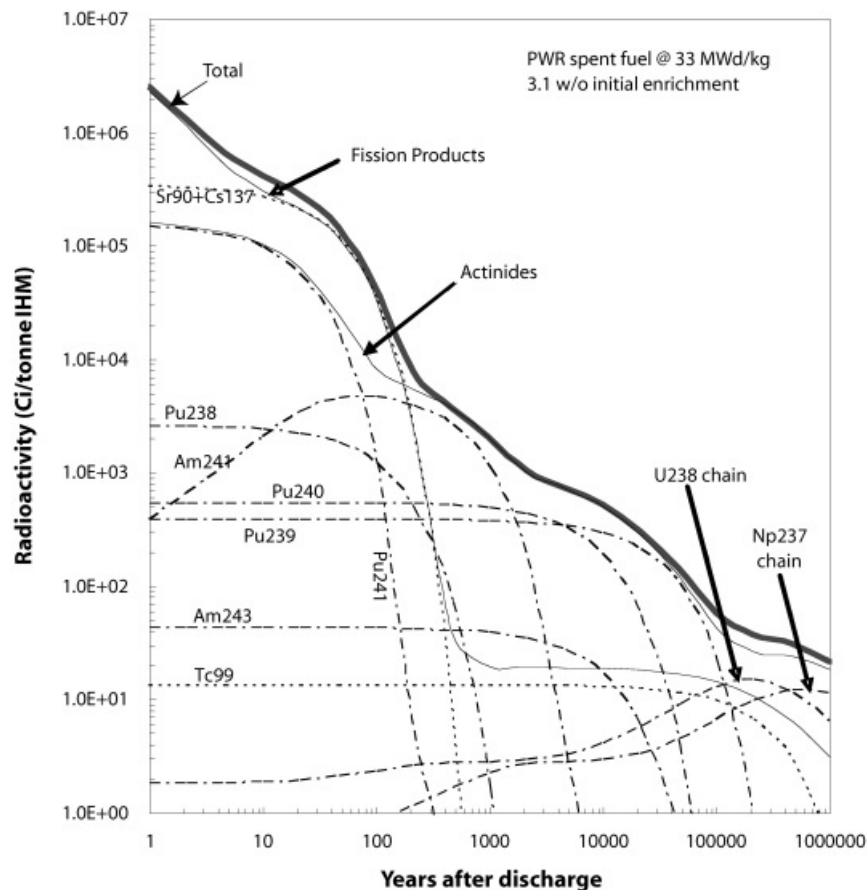


Fig. 1. In situ radioactivity for PWR spent fuel at 33 MWd/kg.

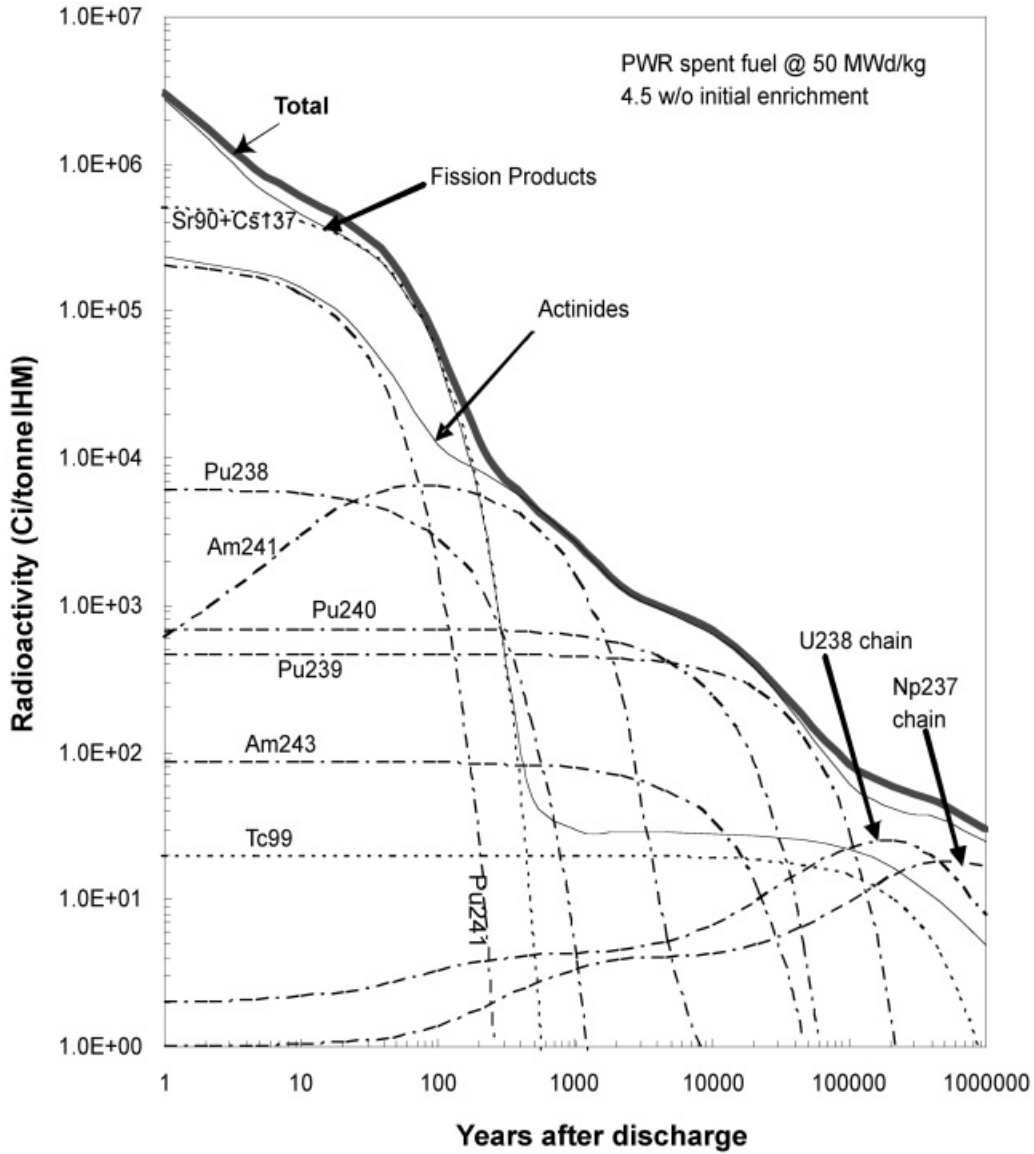


Fig. 2. In situ radioactivity for PWR spent fuel at 50 MWd/kg.

more representative of current PWR fuel discharge burnup. The two graphs show great resemblance in shape, except for their different absolute values. These figures are in units of curies per tonne initial heavy metal (IHM), i.e., based on the same amount of spent fuel. In later discussions, two comparison bases will be considered: per tonne IHM and per gigawatt(electric) year. The first comparison is based on the unit mass of the IHM loading. Given the same 17×17 -lattice assembly design, each assembly has the same fuel loading although the enrichment will be different in order to achieve different burnup levels. The second comparison is based on the unit energy produced in the reactor core. This reflects the amount of spent-fuel generation per unit time of re-

actor operation at a constant power level. If X_1 is per tonne IHM and X_2 is per gigawatt(electric) year, the transformation between the two bases can be expressed as

$$X_2 = \frac{365}{\eta B_d} X_1, \tag{1}$$

where

η = plant thermal to electrical conversion efficiency, i.e., $1150 \text{ MW}(\text{electric})/3411 \text{ MW}(\text{thermal}) = 33.7\%$

B_d = discharge burnup [MWd/kg IHM (same as GWd/tonne IHM)].

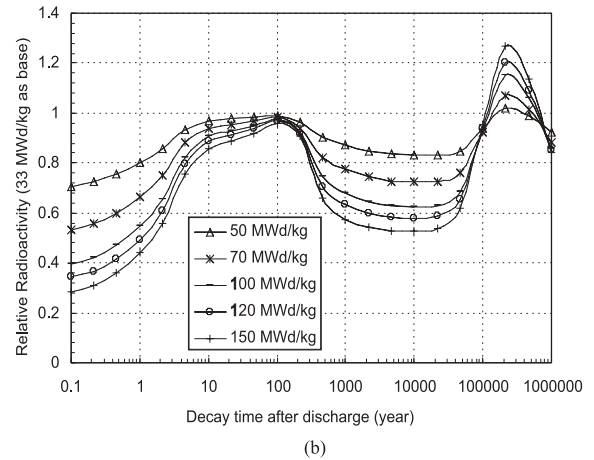
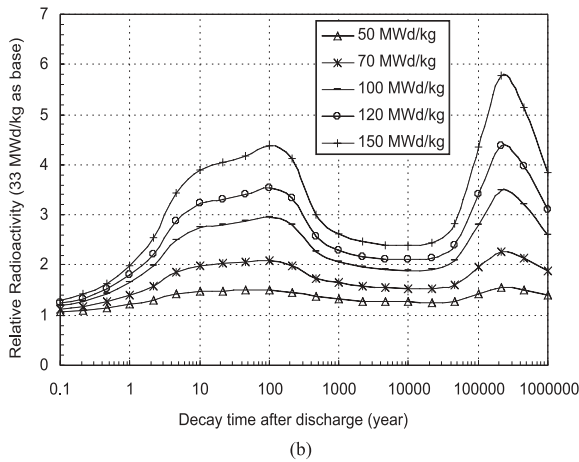
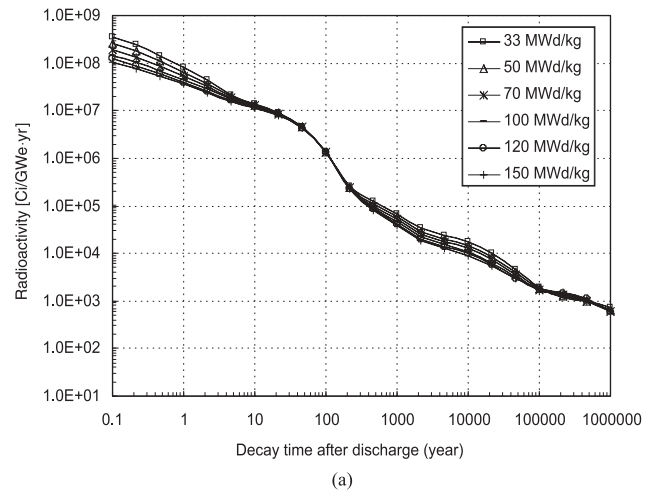
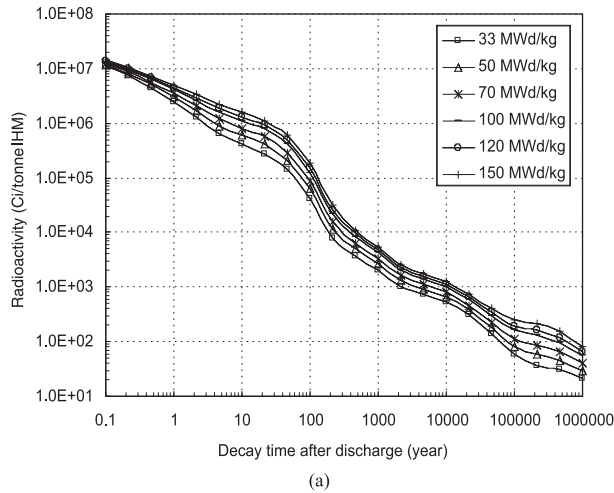


Fig. 3. Radioactivity per tonne IHM versus time after discharge: (a) absolute magnitude and (b) relative magnitude.

Fig. 4. Radioactivity per gigawatt(electric) year versus time after discharge: (a) absolute magnitude and (b) relative magnitude.

The value per unit mass of IHM is a local measure giving the individual assembly characteristics. It is related to the local limits for spent-fuel assemblies, which determine the repository capacity given a fixed volume. The value based on per unit electricity generation is a global measure quantifying the overall spent-fuel characteristics, which indicates the overall repository requirement. Hence, it is directly proportional to the total repository cost.

III.B. Radioactivity and Heat Load

Spent-fuel radioactivity is obviously an interesting quantity. Radioactivity after discharge is shown in Figs. 3 and 4. As shown in Fig. 3, the higher the discharge burnup, the more radioactive is the spent fuel. It is interesting to note from Fig. 4 that in the period from 20 to 200 yr after discharge, the radioactivity per unit electricity is relatively independent of the discharge burnup. However, in the period before 20 yr and after 200 yr, the radioactivity

generated in the spent fuel per unit electricity production clearly favors going to higher burnup.

The radioactivity is a useful metric related to shielding design. Yet, another important quantity is the decay heat, which is of concern in storage facilities and an engineering limit of the waste repository. For example, the currently used preliminary thermal limits in the Yucca Mountain project include (a) the power generation rate of a single waste package (kilowatts) at the time of emplacement and (b) the power generation rate of a line of closely spaced waste packages (in kilowatts per meter) at the time of emplacement.¹¹ The temperatures before closure are controlled by ventilation, and the controlling thermal limits are based on postclosure performance. There are also various temperature limits existing at different locations; e.g., the spent-fuel cladding and the drift rock temperature must not exceed certain values. Obviously, the high-burnup fuel produces more heat; thus, more cooling is required. Given the same waste package design, fewer high-burnup spent-fuel assemblies can be

accommodated in a single waste package. However, there are also fewer spent-fuel assemblies assuming a fixed electricity generation. The overall required repository volume will depend on the total amount of decay power. On the other hand, the total amount of decay heat to be removed is another indicator of the repository cost.

Figures 5 and 6 show the instantaneous decay heat generation rate after discharge. It is seen as expected from Fig. 5 that higher burnup also leads to a higher decay heat generation rate per tonne IHM (or per assembly if the IHM loading is the same). But, based on the unit electricity generation (Fig. 6), the decay power of high-burnup spent fuel is larger than the reference fuel within only two time periods: (a) from 4 to 150 yr and (b) from 150 000 yr to 700 000 yr. Note that the long time frame difference is probably not a concern since (a) significant uncertainties, including both the calculations and the future environment, exist and (b) the decay power is about 5 orders of magnitude lower than in the short term. The short time frame difference suggests that high-burnup spent fuel needs a longer time in storage

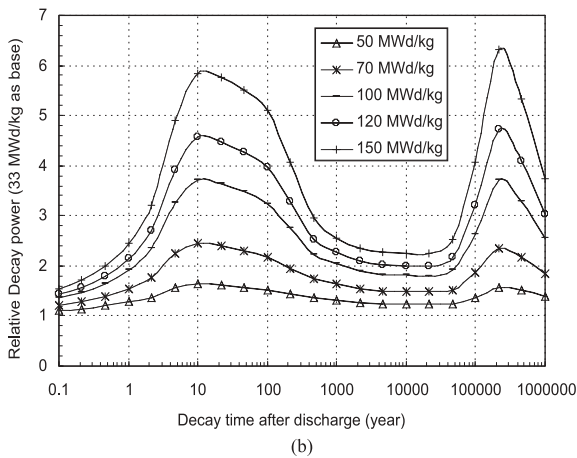
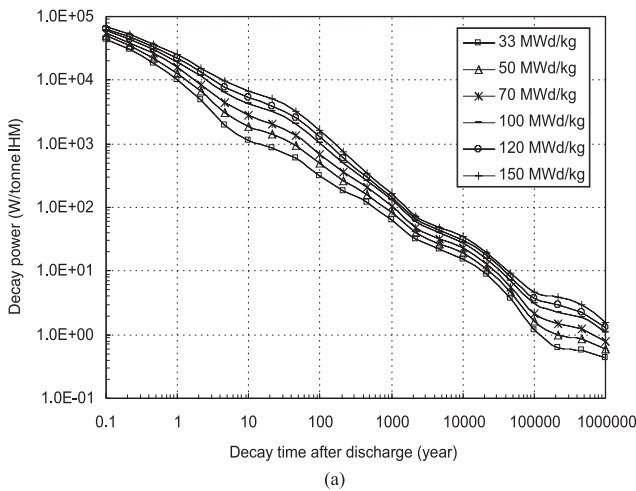


Fig. 5. Decay power per tonne IHM after discharge: (a) absolute magnitude and (b) relative magnitude.

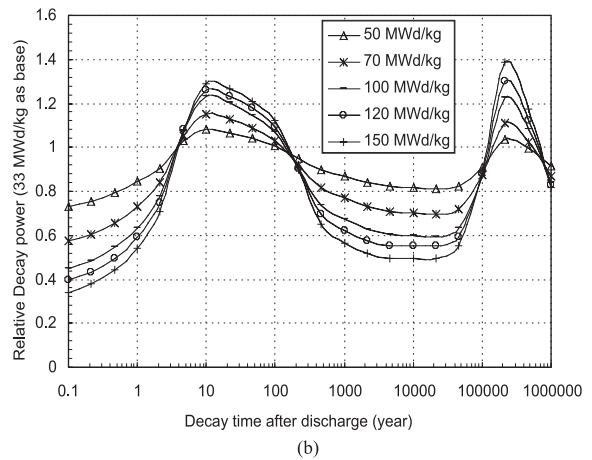
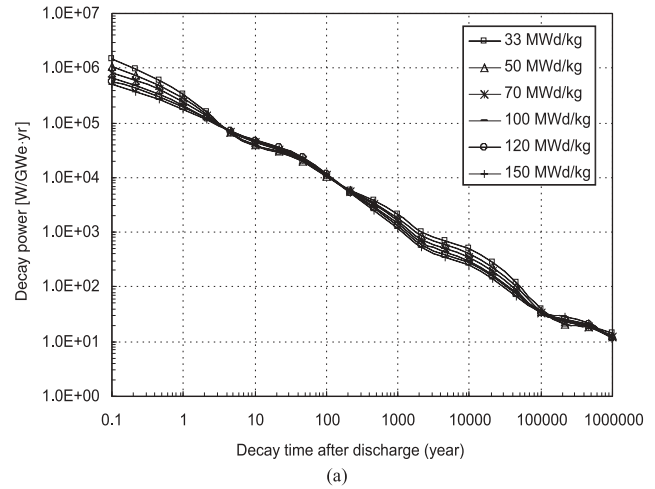


Fig. 6. Decay power per gigawatt(electric) year after discharge: (a) absolute magnitude and (b) relative magnitude.

before final disposal. Per assembly, the high-burnup spent fuel will be hotter (hence a higher surface heat flux), thus more expensive to handle, i.e., fewer high-burnup assemblies in the original cask or the same number of high-burnup assemblies in a newly designed more expensive cask. Figure 6 shows that 150 yr after discharge, the high-burnup fuel would require smaller repository volume while satisfying the same thermal limits. As a matter of fact, comparing Figs. 4 and 6 suggests that the decay power is more challenging than the radioactivity since the functional dependence on burnup is more than linear for the decay power between 4 and 150 yr after discharge.

Next, the accumulated decay energy $E(t)$ is considered, which is defined as the integrated decay power $P(t)$ since emplacement:

$$E(t) = \int_{t_i}^t P(\tau) d\tau, \quad (2)$$

where t_i is time of emplacement after discharge. Since the typical fuel disposal repository design is heat load

limited, i.e., the cost depends on the total amount of heat to be removed, this quantity actually cumulates with the repository cost. For high-burnup cases and assuming immediate emplacement after discharge, the heat load from a single assembly will be higher (Fig. 7); however, fewer assemblies will be discharged per unit energy production. Figure 8 confirms that the total decay heat generation is reduced as burnup increases. Hence, per unit energy production, the high-burnup approach has certain advantages since the accumulated decay heat is reduced. Note that the accumulated decay energy defined here includes all decay energy after discharge from the core. If the time window of 4 to 150 yr is excluded for the repository (Fig. 6), further reduction in the decay energy can be achieved for high-burnup fuel.

Figure 9 reveals a correlation between decay power and radioactivity. Roughly speaking these two quantities are proportional, but at quite different ratios below and above 100 yr, reflecting the isotopic changes. However, at ~10 yr, there are moderate differences among the dif-

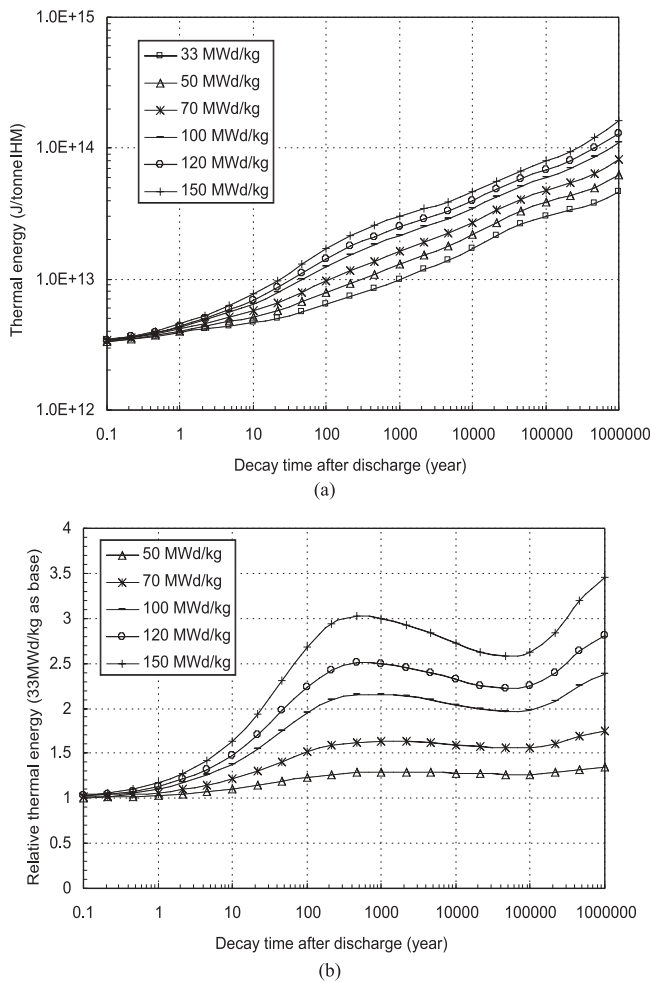


Fig. 7. Accumulated decay heat per tonne IHM after discharge: (a) absolute magnitude and (b) relative magnitude.

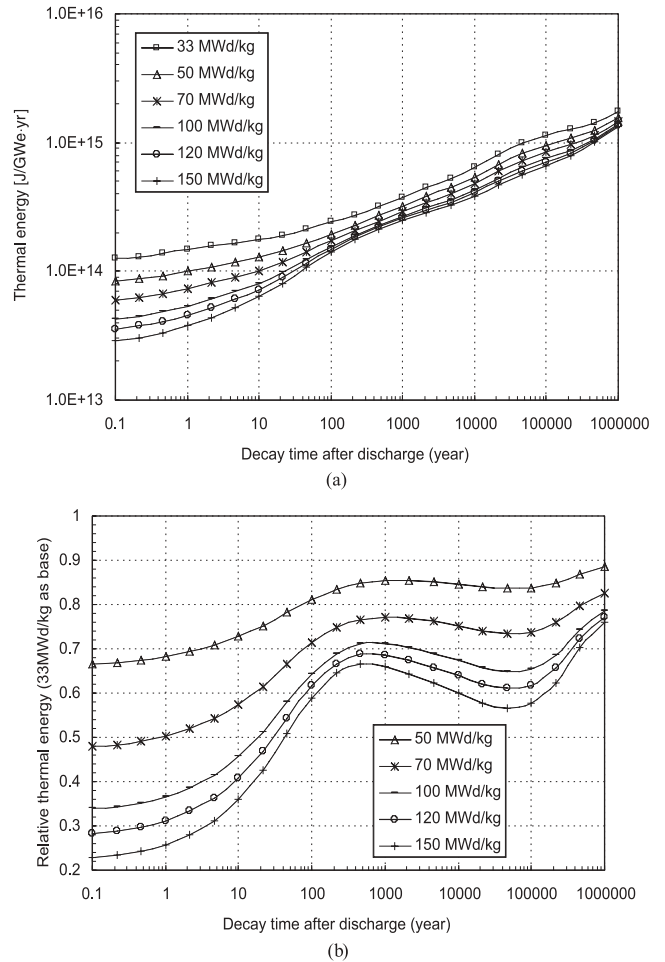


Fig. 8. Accumulated decay heat per gigawatt(electric) year after discharge: (a) absolute magnitude and (b) relative magnitude.

ferent cases. This explains the difference in decay heat power versus time: The radioactivity decreases very fast in the time frame of 100 to 1000 yr after discharge because the fission products decay out. Similarly, Fig. 10 shows the correlation between radioactivity and decay energy. Since the thermal energy increases with time, the ratio between the thermal energy and the radioactivity also increases as a function of time.

A specific example is given in Table II considering an assembly with 100 MWd/kg versus one with 50 MWd/kg burnup, hence the need to handle two assemblies versus one assembly for storage, transportation, and disposal for the same electricity generation. Certain criteria depend more on the value per assembly (local effect); others depend on the total (global effect). For example, the heat flux for cooling is per assembly as is the radioactivity for handling, but cask capacity is more dependent on total decay power for cooling and total radioactivity for shielding. Hence, high burnup complicates spent-fuel handling before it gets to the repository if the same storage time is

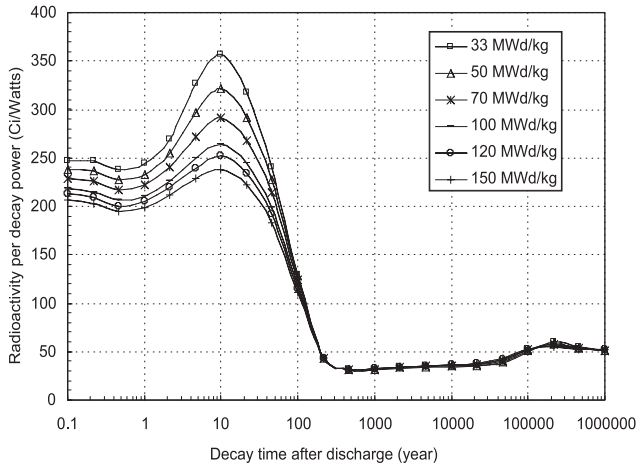


Fig. 9. Correlation between decay power and radioactivity.

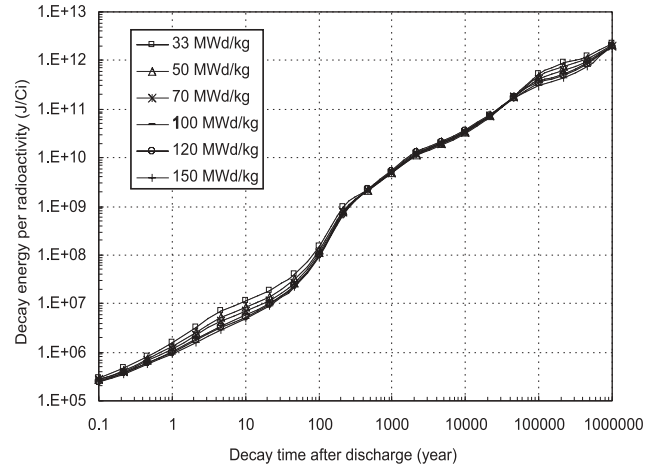


Fig. 10. Correlation between decay energy and radioactivity.

assumed. The heat load between 100 and 1000 yr in the repository clearly shows the benefits of high-burnup fuel, i.e., ~10% savings for the 100 MWd/kg fuel compared to the 50 MWd/kg fuel. In fact, if more fuel storage time is allowed, larger repository savings can be realized for the high-burnup fuel.

III.C. Spent-Fuel Isotopics

As mentioned earlier, only actinides and fission products are considered important in spent fuel. The fraction of fission products in spent fuel is roughly proportional

to the discharge burnup. For example, doubling burnup implies doubling of fission product contents. Under the same three-batch fuel management, the reactivity of spent fuel decreases as burnup increases based on the linear reactivity model.¹² However, per unit of electricity generation, the total amount of fission products produced would be the same regardless of burnup levels. Meanwhile, the fuel burnup has a larger impact on actinides. Generally, LWR depletion results in burning of fissile materials, such as ²³⁵U and newly bred ²³⁹Pu, and accumulation of plutonium and other transuranic elements. Some nuclides reach equilibrium values; e.g., ²³⁹Pu

TABLE II
Comparison of 100 and 50 MWd/kg Spent Fuel Parameters

	Local Effect	Global Effect
	One assembly at 100 MWd/kg One assembly at 50 MWd/kg	One assembly at 100 MWd/kg Two assemblies at 50 MWd/kg
Storage (at 1 yr)		
Radioactivity	1.36	0.68
Decay power	1.51	0.75
Transportation (at 10 yr)		
Radioactivity	1.87	0.94
Decay power	2.28	1.14
Disposal (at 100 yr)		
Radioactivity	1.97	0.98
Decay power	2.12	1.06
Heat load (10 to 100 yr) ^a	2.22	1.11
Repository (at 1000 yr)		
Radioactivity	1.56	0.78
Decay power	1.54	0.77
Heat load (100 to 1000 yr)	1.79	0.89

^aCalculated as the integration of decay power between the interval of 10 to 100 yr following discharge.

saturates at ~ 45 MWd/kg. Most others do not. For example, ^{235}U decreases monotonically, and several minor actinides increase gradually. For the same electricity generation, spent-fuel reduction via high burnup implies reduction of total actinides. From this perspective, the current waste fee based on the electricity generation (or on fission product production) is not fair because it neglects the credit of actinide reduction in the spent fuel (as shown in Table III).

An important issue is criticality control, which depends on the specific spent-fuel waste package design, the geometric configuration, and the nuclide compositions. Clearly, criticality in a repository needs to be

avoided because it might accelerate long-term radionuclide releases to the environment. Criticality safety requires that $k_{eff} \leq 0.95$ at any location. It was proposed in Ref. 13 that depleted uranium dioxide particles made from enrichment plant tails can be added in the LWR waste package for long-term repository criticality control. Because the higher-burnup spent fuel contains more fissile materials, it poses a higher risk for recriticality, especially following the decay of fission products.

The actinides of the spent fuel at different discharge burnups are examined in this section. Since some actinides have relatively short half-lives, such as 13.2 yr for ^{241}Pu , the isotopics are compared at 10 and 1000 yr after

TABLE III
Actinide Compositions in High-Burnup Spent Fuels*

B_d	33 MWd/kg	50 MWd/kg	100 MWd/kg	150 MWd/kg
10 yr after discharge				
^{234}U	0.02	0.02	0.04	0.06
^{235}U	0.90	1.07	1.62	2.21
^{236}U	0.38	0.61	1.47	2.60
^{238}U	97.53	96.76	94.35	91.64
^{237}Np	0.04	0.08	0.20	0.34
^{238}Pu	0.01	0.03	0.14	0.29
^{239}Pu	0.65	0.78	1.12	1.42
^{240}Pu	0.24	0.31	0.46	0.58
^{241}Pu	0.10	0.13	0.22	0.29
^{242}Pu	0.05	0.08	0.15	0.19
^{241}Am	0.06	0.09	0.16	0.21
^{242m}Am	—	—	—	0.01
^{243}Am	0.01	0.02	0.05	0.08
^{244}Cm	—	0.01	0.03	0.05
^{245}Cm	—	—	0.01	0.01
$^{239}\text{Pu}/(\text{Pu}_{total})$	61%	58%	54%	51%
Total Pu (g/MWd)	0.307	0.253	0.187	0.157
Total transuranics (g/MWd)	0.343	0.290	0.226	0.196
Total actinide (g/MWd)	29.3	19.0	8.97	5.63
1000 yr after discharge				
^{234}U	0.03	0.06	0.17	0.35
^{235}U	0.92	1.09	1.65	2.25
^{236}U	0.41	0.64	1.52	2.66
^{238}U	97.54	96.77	94.36	91.65
^{237}Np	0.17	0.25	0.49	0.74
^{238}Pu	—	—	—	—
^{239}Pu	0.63	0.76	1.09	1.39
^{240}Pu	0.22	0.28	0.44	0.57
^{241}Pu	—	—	—	—
^{242}Pu	0.05	0.08	0.15	0.19
^{241}Am	0.03	0.05	0.08	0.11
^{242m}Am	—	—	—	—
^{243}Am	0.01	0.02	0.05	0.07
^{244}Cm	—	—	—	—
^{245}Cm	—	—	—	0.01
$^{239}\text{Pu}/(\text{Pu}_{total})$	70%	68%	65%	65%

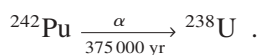
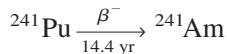
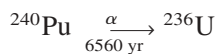
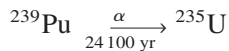
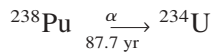
*The actinide compositions are in weight percent.

discharge (Table III). There are more minor actinides generated as the discharge burnup increases. The residual ^{235}U enrichment in the spent fuel increases with burnup. However, ^{236}U also increases significantly. In an LWR the burnup-reactivity worth of ^{236}U is approximately -0.24 relative to ^{235}U at $+1.0$. Therefore, the spent-fuel fissile uranium is worth less than the fissile uranium in the fresh fuel. This feature could be beneficial when considering the repository criticality issue. But, on the other hand, the degraded uranium discourages reprocessing and recycling of uranium in the high-burnup fuel. However, enrichment using gaseous diffusion or centrifuge technology will decrease the $^{236}\text{U}/^{235}\text{U}$ ratio by another factor of 3 or so.¹⁴ The irradiation of ^{236}U produces ^{237}Np , which is the leading dose contributor between 100 000 and one million yr for waste disposal in the Yucca Mountain assessment. Hence, the recycling of uranium in thermal reactors for further irradiation appears to be undesirable from the long-term radiotoxicity point of view.

IV. PROLIFERATION RESISTANCE

IV.A. Introduction

Plutonium is a sensitive nuclear-weapon material. Its content in the spent fuel poses a certain degree of proliferation potential. Fifteen isotopes of plutonium are known; all are radioactive. The principal ones of interest to the design of nuclear weapons, and their half-lives/decay modes, are



Based on technical studies, it is generally agreed that $<20\%$ ^{235}U enrichment in ^{238}U and $<12\%$ ^{233}U enrichment in ^{238}U are not weapons-usable materials.¹⁵ However, unlike uranium, any isotopic mix of plutonium has a relatively small critical mass (~ 10 kg for a bare metallic sphere configuration). Hence, there is no general isotopic concentration threshold for plutonium isotopes from a critical mass point of view. Nevertheless, the suitability for weapons usage varies significantly for plutonium isotopes. In reality, uranium (mostly ^{238}U) is the dominant element in the spent fuel, whereas the plutonium content is about two orders of magnitude lower. Because

uranium and plutonium both exist invariably as mixtures of isotopes, it would be misleading to consider the non-proliferation problem solely in terms of minimizing quantities of fissionable material. The isotopic dilution, as in the case of spent fuel, constitutes an effective, though not an absolute, barrier to proliferation. Table IV lists the important characteristics of plutonium isotopes.¹⁶ Plutonium-238, ^{240}Pu , and ^{242}Pu have high spontaneous neutron generation, which reduces the bomb yield significantly. Plutonium-238 also has a large decay heat, which further complicates the design of an explosive device. Plutonium-241, although a suitable material for weapons, has a relatively short half-life. If a weapon does include ^{241}Pu initially, over several years or even decades, the reactivity worth will decline with time (note that the daughter nuclide, ^{241}Am , is nonfissile in a thermal spectrum but still fissile in a fast spectrum). In conclusion, ^{239}Pu is the only desired isotope for weapons use, and other isotopes are important through their adverse effects.

IV.B. Plutonium Vector

Increasing the discharge burnup is advantageous for improving proliferation resistance. An updated set of correlations can be found in Ref. 17, which relates the isotopic composition with burnup and enrichment for current-design PWR lattice pin cell calculations based on CASMO-4 data. The total plutonium accumulates approximately as the square root of burnup; hence, less is produced per gigawatt(electric) year at higher burnup. For example, doubling the discharge burnup from 50 to 100 MWd/kg decreases the plutonium content per megawatt day by $\sim 25\%$. The ratio of ^{238}Pu to ^{239}Pu increases approximately as burnup to the 2.5 power, whereas the ratio of ^{240}Pu to ^{239}Pu increases approximately proportional to the burnup.

As an example, the fraction of ^{238}Pu in the plutonium is examined at different burnups. Figure 11 shows

TABLE IV
Plutonium Isotope Properties Important
to Proliferation Resistance*

	Half-Life (yr)	Spontaneous Fission Neutrons (n/kg·s ⁻¹)	Decay Heat (W/kg)	Bare Critical Mass (kg)
^{238}Pu	87.7	2 600 000	560	10
^{239}Pu	24 100	22	1.9	10
^{240}Pu	6 560	910 000	6.8	40
^{241}Pu	14.4	49	4.2	10
^{242}Pu	376 000	1 700 000	0.1	100

*Reference 12.

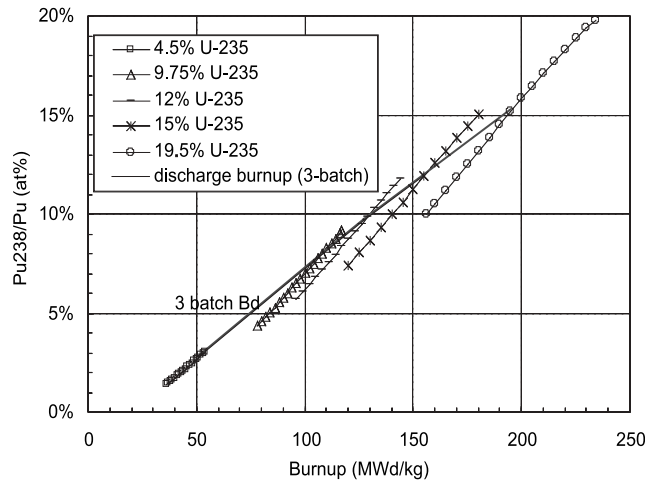


Fig. 11. Plutonium-238 fraction versus burnup for several initial ^{235}U enrichments.

that the fraction of ^{238}Pu in the discharged fuel increases with burnup slightly faster than linearly. This is represented by the intersection of the heavy line showing discharge burnup, with the marked lines showing ^{238}Pu content for various initial enrichments. For example, doubling the burnup (from 50 MWd/kg using 4.5 wt% initial ^{235}U enrichment to 100 MWd/kg using 9.75 wt% ^{235}U enrichment assuming three-batch fuel management) would increase the ^{238}Pu fraction to $\sim 7\%$. Plutonium-238 is so undesirable that some authors see too many difficulties beyond a 2% fraction of the total plutonium.¹⁸ Figure 11 also demonstrates the importance of using an appropriate initial ^{235}U enrichment when calculating ^{238}Pu content. Note that for a fixed discharge burnup (corresponding to a vertical line in Fig. 11), increases in the initial ^{235}U enrichment actually reduce the ^{238}Pu content at discharge. However, the plutonium vector in the spent fuel changes with time because of differences in the decay half-lives among isotopes. In particular, ^{238}Pu and ^{241}Pu have relatively short half-lives; hence, as shown in Table III, the ^{239}Pu fraction in plutonium after 1000 yr is $\geq 65\%$ for all burnup cases. This raises some long-term security concern about the repository.

In conclusion, even assuming that the plutonium can be extracted from the elemental mixture in spent fuel, the isotopic vector of plutonium is less weapons usable at higher burnup, while the total plutonium production is reduced per unit energy generation. However, since any composition of plutonium can be fabricated into a crude, low-yield bomb in the kiloton range given sufficient determination and ingenuity,¹⁶ the impact of the plutonium isotopics may be small. Nevertheless, a recent assessment¹⁹ assigns ^{239}Pu at 40% of total plutonium to a category one step down in weapons usability compared to the 60% representative of current LWR spent fuel; this favors higher burnup.

V. ADDITIONAL CONSIDERATIONS

From a nuclear utility point of view, economic viability is a top priority. The historical burnup increase is driven partially by the fuel cycle cost savings. Assuming a fixed 18-month cycle and representative economic parameters prevailing in 2003, the fuel cycle cost of a discharge burnup ranging between 40 and 100 MWd/kg is shown in Fig. 12 (Ref. 20). The unit cost parameters, such as uranium ore and separative work unit (SWU) costs, are assumed to be the same throughout the entire burnup range even though the fuel enrichment is beyond 5 wt% for burnup above 55 MWd/kg. It can be seen that the fuel cost has a weak dependence on burnup (minimum at ~ 70 MWd/kg). In view of swings in the cost of uranium ore and enrichment services, there is a mild economic incentive for current PWRs to go beyond 50 MWd/kg from the nuclear fuel cycle cost perspective. Similar observations are made from an Electric Power Research Institute study.²¹ The uranium ore purchase and the SWU cost are dominant contributors to the total fuel cycle cost. The spent-fuel storage component is a relatively small contributor. Therefore, the cost-competitiveness of high burnup must originate from other considerations.

There are also technical challenges for increasing burnup. Generally speaking, increased fuel enrichment is needed, which hardens the neutron spectrum. Consequently, the core criticality control is more challenging because of reduced reactivity worth of control materials. In particular, the soluble boron concentration, limited by

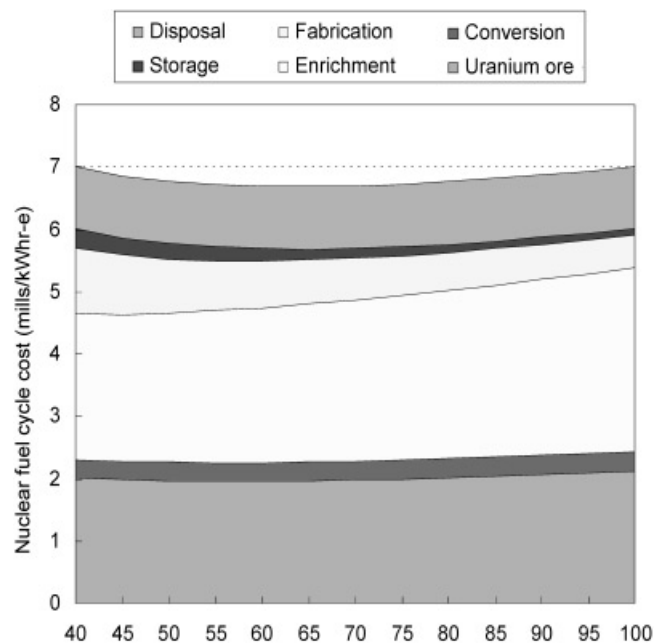


Fig. 12. Fuel cycle cost for 18-month conventional PWRs.

the primary-loop water chemistry, suppresses less excess reactivity. More burnable poison will then be needed. Under a hardened neutron spectrum, it is preferred to distribute the burnable poison in a more homogeneous way because of the reduced poison depletion rate. Relatively weak absorbers, such as erbium and boron, might not be adequate for beginning-of-cycle reactivity control and might not burn completely during the cycle. In addition, the core shutdown margin would also be compromised. The high-burnup core, in some sense, exhibits similar behavior to mixed-oxide cores. There are several ways to overcome the high-burnup core challenges:

1. The obvious one is to increase the amount of control materials in the core, e.g., more control rods and more highly enriched absorbers.
2. The hydrogen-to-heavy-metal ratio can be adjusted in order to recover control materials worth.
3. The fuel management scheme can be adjusted.

Although feasible, the first remedy is costly. The second one is beneficial, but the fuel loading is usually reduced, which penalizes the initial enrichment. The last approach involves fuel management. For example, the number of reload assemblies can be increased, hence alleviating the spectrum hardening. Also, the core power density can be significantly increased using internally and externally cooled annular fuel,²² which is in principle equivalent to cycle length extension at nominal power.

In summary, high-burnup fuel may be desirable from a waste management prospectus, but it would require considerable work on the fuel and core design in order to maintain economic viability. Considerable research and development work is also needed to ensure fuel reliability over a longer in-core exposure. Finally, the back-end high-burnup fuel benefits need to be properly reflected in the waste fee. As discussed in Sec. IV, the current waste fee based on electricity generation can be viewed as a fission product waste fee, whereas the reduction of total actinides is not considered for the high-burnup fuel. And, the decay heat shows a less than linear increase as a function of burnup. Also, it should be recognized that there are other quantities that increase faster than linearly with burnup, e.g., the neutron source in the spent fuel as reported in Ref. 23. The waste fee needs to be properly revised to reflect the actinide reduction credit, perhaps by changing from a per kilowatt(electric) hour to a toxicity basis.

VI. CONCLUSIONS

Examination of several proposed burnup levels shows that high burnup is beneficial in terms of reducing the overall accumulative spent-fuel radiotoxicity and decay energy as well as improving the proliferation resistance,

even while an individual assembly is more radioactive and hotter. This is because the burnup dependence of the spent-fuel radioactivity and decay energy is less than linear. It is found that a longer storage time (~ 150 yr storage) is beneficial for the high-burnup fuel, which can avoid the high decay heat penalty for the high-burnup fuel. To realize benefits for waste disposal, the high-burnup credit needs to be recognized in an appropriate way, such as adjusting the waste fee to a per toxicity basis. The majority of waste fee should also be paid when spent-fuel assemblies are passed to the government's custody.

The implementation of high-burnup fuel would imply a broader scope of work since fuel design changes will be necessary to respond to the challenges of neutron spectrum hardening.

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