APPENDIX C DIFFERENCES BETWEEN THERMAL AND RADIATION PROPERTIES OF MIXED OXIDE AND LOW-ENRICHED URANIUM SPENT NUCLEAR FUEL

This appendix reviews the expected differences between thermal and radiation properties of mixed oxide (MOX) and low-enriched uranium (LEU) spent nuclear fuel (SNF). Limited experimental information is available for MOX SNF, so determining what to expect from various grades of plutonium (see below), assembly types, fuel pellet types, reactor categories, and amount of burnup is determined solely from performing source term calculations. While only limited studies have been performed to understand what might be expected from these types of variations, educated estimates for these differences are attempted here and noted in the text or in footnotes. MOX SNF comes from MOX fresh fuel that has been irradiated in a thermal reactor. Appendix B to this SRP provides information regarding the compositions of MOX fresh fuel (see pages B-1 and B-2).

Oak Ridge National Laboratory (ORNL) conducted a detailed study of the rates of heat generation, gamma emission, and neutron emission due to decay for MOX fuel irradiated in various reactors. The following four ORNL reports present the results for SNF from the reactors stated in the reports' titles:

(1) "Characteristics of Spent Fuel from Plutonium Disposition Reactors, Vol. 1: The Combustion Engineering [CE] System 80+ Pressurized-Water-Reactor Design" (Murphy 1996)—this report gives the results for both MOX fuel and LEU fuel; the assessment given for MOX fuel assemblies and LEU fuel assemblies were used in this appendix for generic fuel comparisons for pressurized-water reactors (PWRs)

(2) "Characteristics of Spent Fuel from Plutonium Disposition Reactors, Volume 2: A General Electric [GE] Boiling-Water-Reactor Design" (Ryman and Hermann 1998)—this report gives the results for both MOX fuel and LEU fuel; the assessment given for MOX and LEU fuel assemblies were used in this appendix for generic fuel comparisons for boiling-water reactors (BWRs)

(3) "Characteristics of Spent Fuel from Plutonium Disposition Reactors, Vol. 3: A Westinghouse Pressurized-Water Reactor Design" (Murphy 1997)

(4) "Characteristics of Spent Fuel from Plutonium Disposition Reactors, Vol. 4: Westinghouse Pressurized-Water-Reactor Fuel Cycle without Integral Absorber" (Murphy 1998)

For each reactor type, it is possible to (i) select from a number of different fuel assemblies, (ii) for MOX, choose different arrangements of fuel pins having different compositions of plutonium, uranium, and burnable absorbers, and (iii) use annular fuel pellets rather than cylindrical fuel pellets. All these changes can affect the total burnup and the amount of heavy metal contained in the MOX fuel or LEU fuel assemblies. The ORNL studies focused on identifying differences in spent fuel characteristics that are significantly greater than typical burnup-related variations. It is expected that increasing the burnup of both MOX fuel and LEU fuel assemblies would result in larger differences in spent fuel characteristics. The first two ORNL reports in the list above (Volumes 1 and 2 of the spent fuel studies) were chosen for this study because they consider typical differences in SNF characteristics, and they are the only

ones available that compare LEU SNF to MOX SNF. However, they do not necessarily represent analyses that give bounding differences in SNF characteristics.

The ORNL studies used weapons grade (WG) plutonium for their MOX fuel rods. The 3013 Standard (DOE, 2012) gives the weight percent (wt%) for various isotopes in various grades of plutonium (see Table B–6 of the 3013 Standard). The ORNL studies used weight percents of various plutonium isotopes consistent with those for WG plutonium listed in Table B–1 of Appendix B to this SRP. Details of the fuel assemblies used in the ORNL studies are presented below.

A discussion of the characteristics of the Combustion Engineering System 80+ PWR (CE-PWR) MOX SNF and PWR LEU SNF fuel assemblies is presented below. For purposes of comparison, these same data are also summarized in Table C–1. Table C–2 shows the irradiation characteristics of the CE System 80+ fuel assemblies. Table C–3 shows a comparison of fuel assembly characteristic of the GE BWR MOX SNF and BWR LEU SNF fuel assemblies. Table C–4 shows the irradiation characteristics of the GE BWR fuel assemblies.

The MOX fuel for the CE-PWR irradiation contained 6.7 wt% WG plutonium and 91.3 wt% depleted uranium,[1] together with 1.9 wt% of erbium, in the form of erbium oxide (Er_2O_3), as components of the heavy metal. The core also contained Al_2O_3-B_4C burnable poison rods (BPRs). The assembly studied, known as the shim assembly, contained a 16×16 square array that was 20.25 centimeters (cm) on a side, with a fuel-rod pitch of 1.29 cm. The assembly studied contained 256 fuel rod positions with a total of 224 fuel rods, four control rods, one instrument tube, and 12 BPRs. The four control rods and single instrument tube displaced the equivalent of 20 fuel rod positions. The assembly contained 0.419 metric tons of heavy metal (MTHM) in the 224 fuel rods, not counting the 1.9 wt% of erbium. The burnup criterion used was 28.9 MW/MTHM, and the assembly was burned to 17,681.8 megawatt days (MWd), in four cycles of 365 days each. A 30-day downtime was allowed between cycles. This represents an assembly power level of 12.34 megawatts (MW), and a burnup of 42.2 gigawatt days per metric ton of heavy metal (GWd/MTHM) (see Table C–1).

The LEU fuel for the CE-PWR irradiation contained 4.2 wt% uranium-235 and 95.8 wt% uranium-238, as components of the heavy metal, in 224 identical fuel rods. In addition, 12 fuel rods contained 4.1 wt% uranium-235 and 94.0 wt% uranium-238, together with 1.9 wt% of erbium, in the form of Er_2O_3, as components of the heavy metal. These 12 fuel rods were located in the same positions where the 12 BPRs were located in the MOX case discussed above. The shim assembly studied contained the same 16×16 square array that was 20.25 cm on a side, with a fuel-rod pitch of 1.29 cm. The assembly studied also contained the same four equivalent control rods and one equivalent instrument tube as the MOX assembly. The assembly contained 0.424 MTHM in the 236 fuel rods, not counting the same 1.9 wt% of erbium. The burnup criterion used was 29.1 MW/MTHM, and the assembly was burned to 20,267.2 MWd, in three cycles of 18 months each. A comparable 30-day downtime was allowed between cycles. This represents an assembly power level of 12.34 MW, which was the same as for the MOX fuel assembly. The burnup was 47.8 GWd/MTHM (see Table C–2).

[1]Depleted uranium is 99.8 wt% uranium-238 and 0.2 wt% uranium-235.

Table C–1 Comparison of fuel-assembly characteristics for the combustion engineering system 80+ pressurized-water-reactor SNF		
Characteristic	**CE-PWR MOX**	**CE-PWR LEU**
Weight heavy metal (MT)	0.419	0.424
wt% WG plutonium	6.7	NA
wt% uranium	91.3 [a]	100 (4.2 ^{235}U)
wt% erbium (Er_2O_3)	1.9	1.9
Burnable poison rod (BPR) material	Al_2O_3-B_4C	NA
Array size	16×16 (20.25 on side)	16×16 (20.25 on side)
Fuel rod pitch (cm)	1.29	1.29
Number of rods	256	256
Fuel rods	224	236
Control rods	4	4 (equivalent)
Instrument tubes	1	1
BPRs	12	NA
Burnup criterion (MW/MTHM)	28.9	29.1
Burnup (MWd)	17,681.8	20,267.2
Cycles/length	4/365 days each	3/18 months each
Assembly power level (MW)	12.34	12.34
Representative burnup (GWd/MTHM)	42.2	47.8
[a]Depleted uranium is 99.8 wt% uranium-238 and 0.2 wt% uranium-235.		

Table C–2 Irradiation characteristics of Combustion engineering System 80+ pressurized-water-reactor SNF			
Fuel Type	**MTHM**	**Irradiation (days)**	**Burnup (GWd/MTHM)**
MOX	0.419	1,460	42.2
LEU	0.424	1,620	47.8

The MOX fuel for the GE-BWR-5 [2] irradiation contained 2.97 wt% WG plutonium, 96.50 wt% depleted uranium, and 0.53 wt% gadolinium, as components of the heavy metal. The assembly studied contained an 8×8 array that was 15.24 cm on a side, with a fuel rod pitch of 4.129 cm. The assembly contained 64 fuel rod positions, with a total of 60 fuel rods and one guide tube. Seven different types of fuel rods were used, each having a different amount of plutonium, uranium, and gadolinium. The guide tube displaced the equivalent of four fuel rod positions. The assembly contained 0.179 MTHM in the 60 fuel rods, not counting the 0.53 wt% of gadolinium. The burnup criterion used was 25.5 MW/MTHM, and the assembly was burned to 6,715.4 MWd, in four cycles of 340-day uptime, and a 113-day downtime, each with an additional final 113-day uptime. This amounted to an assembly power level of 4.610 MW and a burnup of 37.6 GWd/MTHM (see Table C–3).

The LEU fuel for the GE-BWR-5 irradiation contained 3.25 wt% uranium-235 and 96.75 wt% uranium-238, as components of the heavy metal, in 56 identical fuel rods.

[2] The report actually refers to the GE-BWR-5 but used some features of the GE-BWR-9, such as the four water rods.

Table C–3 Comparison of fuel assembly characteristic for the General Electric boiling-water-reactor SNF		
Characteristics	**GE-BWR MOX**	**GE-BWR LEU**
Weight heavy metal (MT)	0.179	0.183
wt% WG plutonium	2.97	NA
wt% uranium	96.50[a]	100 (3.25 ^{235}U)
wt% gadolinium (Gd_2O_3)	0.53	2.17
Array size	8×8 (15.24 cm on side)	8×8 (15.24 cm on side)
Fuel rod pitch (cm)	4.129	4.129
Number of rods	64	64
Fuel rods	60	60
Guide tube	1	1
Burnup criterion (MW/MTHM)	25.5	25.5
Burnup (MWd)	6,715.4	6,880.8
Cycles/length	4/340-day uptime, 113-day downtime, with an additional 113-day uptime, each	4/340-day uptime, 113-day downtime, with an additional 113-day uptime, each
Assembly power level (MW)	4.610	4.724
Representative burnup (GWd/MTHM)	37.6	37.6
[a]Depleted uranium is 99.8 wt% uranium-238 and 0.2 wt% uranium-235.		

In addition, four fuel rods with burnable absorbers were used, containing 2.17 wt% of Gd_2O_3. The assembly studied contained the same 8×8 array that was 15.24 cm on a side, with a fuel rod pitch of 4.129 cm. The assembly studied contained a total of 60 fuel rods and one guide tube. The assembly contained 0.183 MTHM in the 60 fuel rods, not counting the 2.17 wt% of gadolinium. The burnup criterion used was 25.5 MW/MTHM, which was the same as for the MOX fuel assembly. The assembly was burned to 6,880.8 MWd, in four cycles of 340-day uptime, and a 113-day downtime, each with an additional final 113-day uptime. This amounted to an assembly power level of 4.724 MW, and an identical burnup of 37.6 GWd/MTHM (see Table C–4).

The ratios for heat generation rates, photon emission rates, and neutron emission rates vs. time-from-discharge for the CE-PWR fuel assemblies are shown below in Figures C–1, C–2, and C–3, respectively. The data presented in these figures were calculated by taking the calculated rates of heat generation, gamma emission, and neutron emission due to decay for the MOX fuel assembly irradiation and dividing them by the similar quantities for the LEU fuel assembly. The differences in calculated decay rates for these quantities for the MOX fuel assembly irradiation and the LEU fuel assembly irradiation in a PWR are attributed primarily to differences in fuel material for the purposes of this study.

The ratios for heat generation rates, photon emission rates, and neutron emission rates vs. time-from-discharge for the GE BWR fuel assemblies are also shown in Figures C–1, C–2, and C–3, respectively. Again, the data presented in these figures were calculated by taking the calculated rates of heat generation, gamma emission, and neutron emission due to decay for the MOX fuel assembly irradiation and dividing these by the similar quantities for the LEU fuel assembly. And, again, the differences in calculated decay rates for these quantities for the MOX fuel assembly irradiation and the LEU fuel assembly irradiation in a BWR are attributed primarily to differences in fuel material for the purposes of this study.

Table C–4 Irradiation characteristics of General Electric boiling-water-reactor SNF			
Fuel Type	**MTHM**	**Irradiation (days)**	**Burnup (GWd/MTHM)**
MOX	0.1786	1,473	37.6
LEU	0.183	1,473	37.6

Figure C–1 for heat generation rate shows that the heat rate generated by the MOX SNF and LEU SNF is within about 15 percent of each other over a period of 10 years after discharge. Figure C–2 for decay gamma emission rate, where only gamma energies greater than 250 kilo electron volts (keV) are included in the curves,[3] shows that the decay gamma emission rate generated by the MOX SNF and LEU SNF are also within about 15 percent of each other over a period of 10 years after discharge. Figure C–3 for decay neutron emission rate shows that the decay neutron emission rate generated by the MOX SNF and LEU SNF differs by up to about a factor of 2.5 over a period of 10 years after discharge.[4] These results are based on a single assembly type and fuel composition for each of the two categories of reactors studied. WG plutonium was used for both studies.

Most of the benchmarking that ORNL has investigated for decay heat and radiation source terms has involved LEU fuel. Limited MOX benchmarks indicate that the predicted actinide concentrations, particularly the fissile plutonium isotopes and many fission products, are not nearly as accurate for MOX fuels as previously observed for commercial LEU fuels. For example, plutonium-239 tends to be over-predicted by about 10 to 50 percent, and americium isotopes are also significantly over-predicted by about 25 percent. The reasons for this are not entirely clear, but it could be due to larger uncertainties in the plutonium and other higher actinide cross sections (compared to uranium) that are more important in MOX fuel, and/or the more heterogeneous MOX cores (i.e., when MOX assemblies with different heavy metal compositions are irradiated together with LEU assemblies). It is difficult to know the accuracy of decay heat predictions based on these results, but in general, it is expected that at longer cooling times where actinides dominate, code predictions may overestimate decay heat by potentially 10–20 percent or more for MOX SNF based on the calculated plutonium and americium nuclide inventories. However, several dominant decay heat nuclides important at shorter cooling times are significantly under-predicted (Murphy and Primm 2000).

The accuracy of MOX decay heat calculations would apparently be much lower than for LEU fuels, but it may be conservative for longer cooling times and nonconservative for short cooling times. For neutron source terms, comparisons with the limited benchmark data indicate that SCALE (ORNL, 1995) predictions are in very good agreement for MOX fuel in a PWR but are over-predicted (~20 percent) for MOX fuel in a BWR (Gauld 2002). Uncertainties in the computational predictions by the amounts estimated above (10–20 percent) support the differences in the values shown in Figures C–1 through C–3.

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[3] The shielding associated with SNF packagings is expected to absorb essentially all gammas with energies less than 250 keV.

[4] The curves for each of the figures are based on different fuels and different burnups. While these differences affect the curves shown in Figures C–1 and C–2, the effects are more noticeable in Figure C–3. This is due to the greater sensitivity of the neutron source to differences between the MOX and LEU assemblies and their irradiation for the two analyzed PWR and BWR assembly types. Thus, to understand the figures, particularly Figure C–3, the differences in the fuels and their burnups, including the influence of the fuel property differences to either magnify or minimize reactor operation characteristics (e.g., void fraction in a BWR), need to be considered.

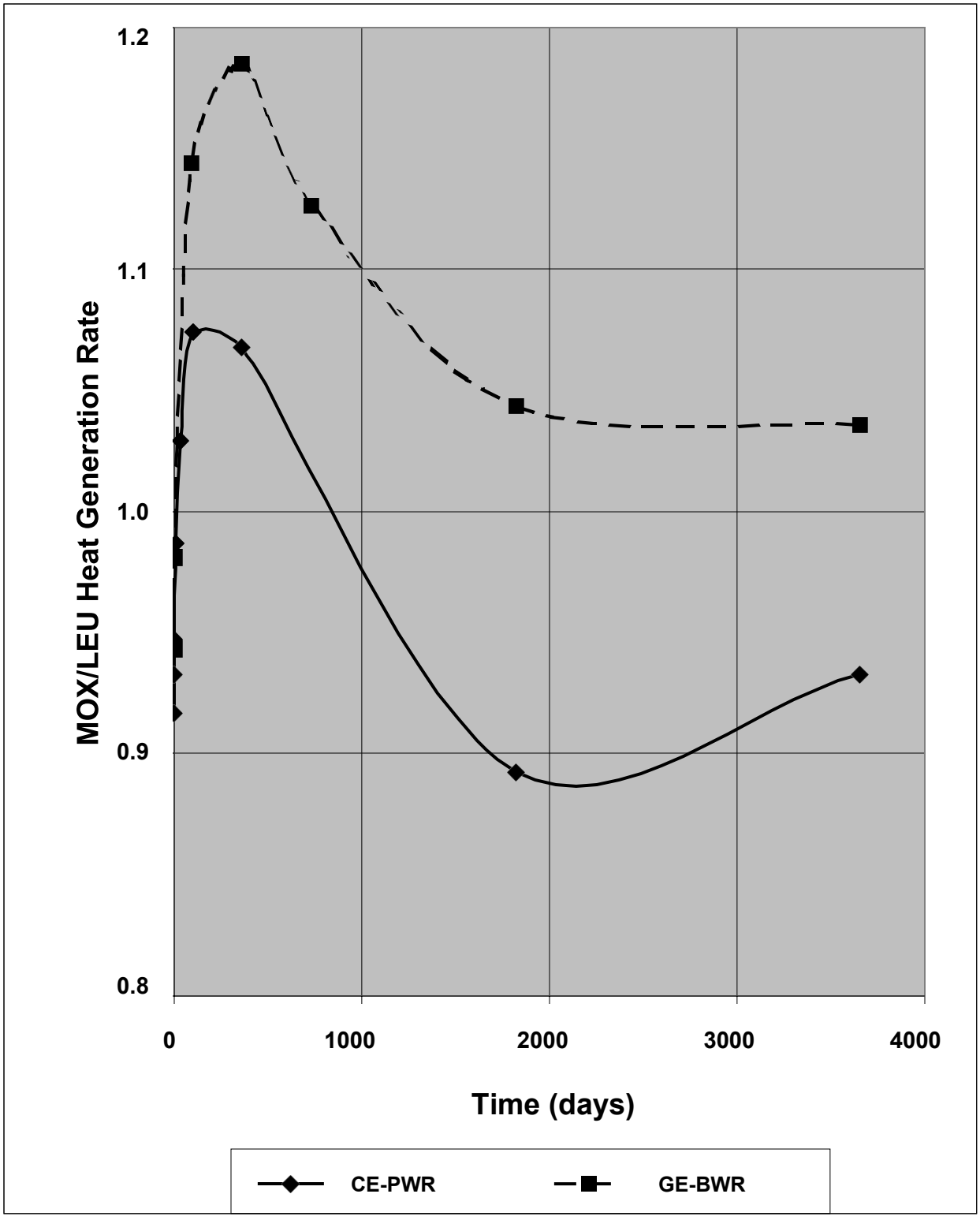

Figure C–1 Ratio of MOX to LEU decay heat generation rate vs. time-from-discharge for Combustion Engineering System 80+ Pressurized Water Reactor (CE-PWR) and General Electric Boiling-Water Reactor Model 5 (GE-BWR-5)

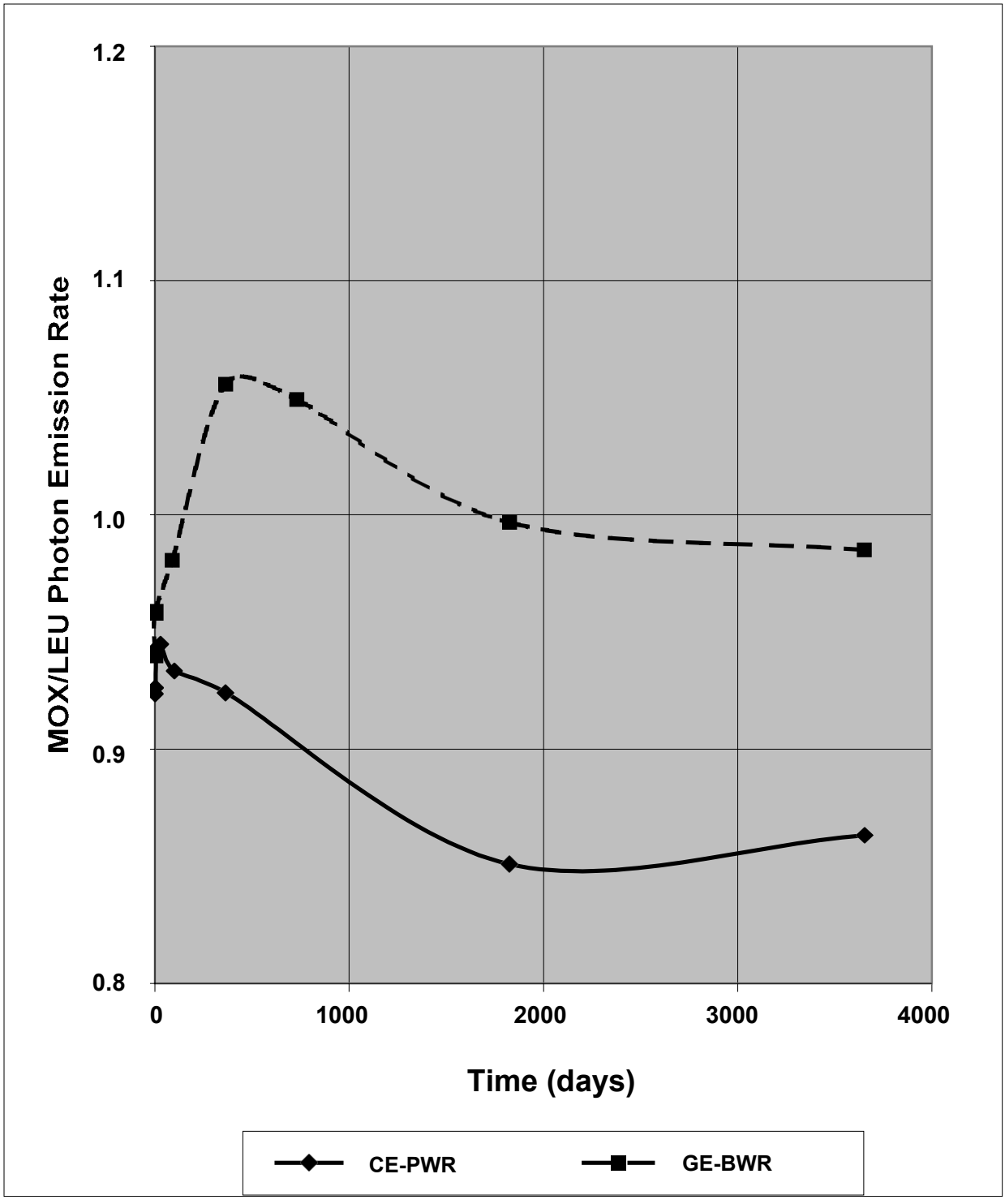

Figure C–2 Ratio of MOX to LEU decay gamma emission rate vs. time-from-discharge for Combustion Engineering System 80+ Pressurized-Water Reactor (CE-PWR) and General Electric Boiling-Water Reactor Model 5 (GE-BWR)

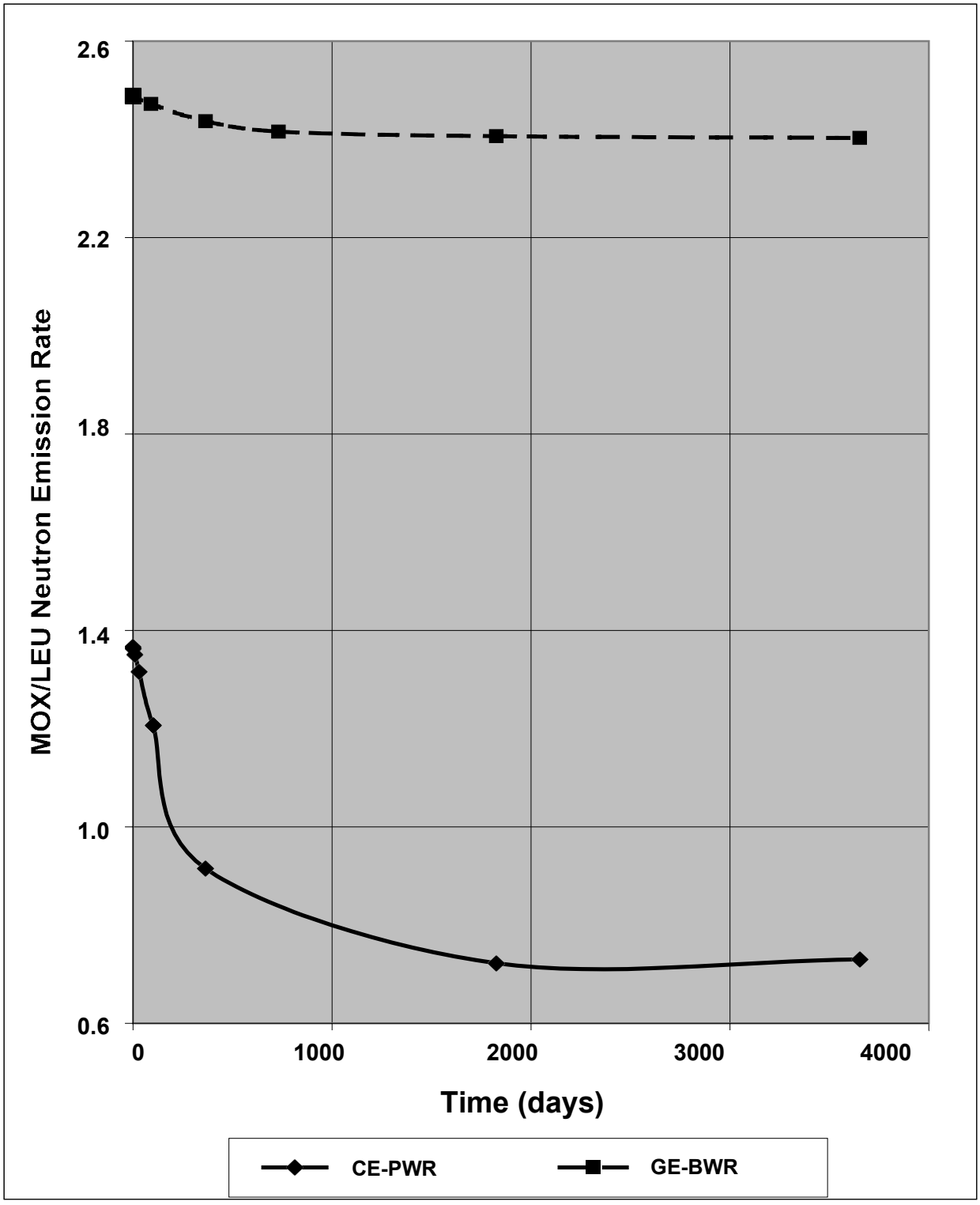

Figure C–3 Ratio of MOX to LEU decay neutron emission rate vs. time-from-discharge for Combustion Engineering System 80+ Pressurized-Water Reactor (CE-PWR) and General Electric Boiling-Water Reactor Model 5 (GE-BWR)

The use of ENDF/B-IV and earlier cross sections have the effect of over-predicting the multiplication coefficient, k_{eff}, for materials containing plutonium. The use of newer, and presumably more accurate, ENDF/B-V and VI cross sections do a better job of predicting k_{eff}. However, the effect of newer cross sections will not necessarily be less conservative for calculating decay heat and radiation source terms when compared to the earlier ones. The much larger isotopic biases observed for MOX fuels in limited benchmark studies are likely to translate into higher uncertainties (biases) in aggregate fuel properties; they may translate to a lesser extent than the isotopic analyses might suggest due to cancellation of errors [e.g., bulk fuel properties are generally predicted better than individual isotopic analyses (Gauld 2002)].

Are the studies shown in Figures C-1 through C-3 representative of other assembly types, fuel pellet types, reactor categories, and burnups that might be considered? The decay heat emission rate and gamma emission rate using WG plutonium are expected to be similar. That is, quantities of heat emission rate and gamma emission rate for MOX SNF and LEU SNF should be roughly within the same envelope determined in the ORNL studies (i.e., within about 40 percent of each other over a period of 10 years after discharge, including benchmark and cross section uncertainties).[5] Using WG plutonium, we estimate (since no systematic studies have been performed as yet) that the decay neutron emission rate for MOX SNF may be up to a factor of 4 larger than that for LEU SNF over a period of 10 years after discharge, taking into account benchmark and cross section uncertainties.[6] The uncertainties are not expected to apply to shorter cooling times, relative to a discharge time of 10 years. The differences, as always, need to be confirmed by independent verification using established radiation transport codes and cross-section sets.

Will these relationships change when studies are made with MOX fuel produced with fuel grade or power grade plutonium? The answers for heat generation and gamma emission rates due to decay are expected to be similar but differ by a larger amount. The use of plutonium containing less plutonium-239 and more of other plutonium isotopes means larger masses of plutonium might be required in the fuel rods, which increases the amount of other isotopes of plutonium in MOX fresh fuel. Irradiation of fuel rods containing more of the other plutonium isotopes is expected to generate a greater heat generation rate and to emit a greater decay gamma emission rate than the WG plutonium used in the MOX fuel studied in the ORNL reports. The additional amount of other plutonium isotopes is expected to generate greater heat generation and gamma emission rates due to decay after irradiation. The heat generation and gamma emission rates due to decay for MOX SNF and LEU SNF might be within about 100 percent of one another over a period of 10 years after discharge, including benchmark and cross-section uncertainties, although without systematic studies this is just an estimate. The uncertainties are not expected to apply to short cooling times relative to a time after discharge of 10 years.

For decay neutron emission rates, it may be more difficult to determine the amount of increase that might be expected with MOX fuel produced with another grade of plutonium, since no systematic studies have been performed as yet. The decay neutron emission rates from other grades of plutonium can be two to four times larger than those for WG plutonium. Again, the use of plutonium containing less plutonium-239 and more of other plutonium isotopes means larger masses of plutonium might be required in the fuel rods, which increases the amount of the other isotopes of plutonium in MOX fresh fuel. Irradiation of fuel rods containing more of

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[5] This is just an opinion, since the uncertainty may be larger than the increase estimated by 20 percent × 2 = 40 percent.

[6] This is also just an opinion, since the uncertainty may be larger than the increase estimated by $2.5 \times 1.5 \cong 4$ factor.

other plutonium isotopes is expected to generate a greater decay neutron emission rate than the WG plutonium used in the MOX fuel studied in the ORNL reports. MOX fuel produced with power grade plutonium has considerably more plutonium-241 present in the fresh fuel. Americium-241 is produced by beta decay of plutonium-241 with a half-life of 14.4 years. For times after discharge less than a year, neutrons from curium-242 and curium-244 can predominate after discharge for several months or so, after which the neutrons from curium-242 decrease significantly. Neutrons from plutonium-240 and americium-241 may also become significant. The neutron emission rates for MOX SNF and LEU SNF should be within an order of magnitude of one another over a period of 10 years after discharge, including benchmark and cross-section uncertainties, although without systematic studies this is just an estimate. The uncertainties are not expected to apply to short cooling times relative to a time after discharge of 10 years. The differences, as always, need to be confirmed by independent verification using established radiation transport codes.

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