APPENDIX B DIFFERENCES BETWEEN THERMAL AND RADIATION PROPERTIES OF MIXED OXIDE AND LOW-ENRICHED URANIUM RADIOACTIVE MATERIALS

The contents considered in this Standard Review Plan (SRP) appendix are unirradiated mixed oxide (MOX) radioactive material (RAM), in the form of powder, pellets, fresh fuel rods, or fresh reactor fuel assemblies. Unirradiated MOX RAM will also be referred to in this appendix as MOX fresh fuel. This appendix summarizes the relative degree of differences between the thermal and radiation properties of the various MOX RAM contents relative to similar properties for analogous low-enriched uranium (LEU) RAM contents. MOX fresh fuel can be made with plutonium having various compositions of plutonium isotopes. The discussion in this appendix makes use of the 3013 Standard (DOE 2012), which specifies the typical grades of plutonium that are used to make the MOX fresh fuel. The actual plutonium compositions found in practice may not match these compositions exactly, but these grades can be considered typical for the purposes of this appendix.

Table B–6 of the 3013 Standard gives weight percents for various plutonium isotopes in various grades of plutonium. They are reproduced in the following table (Table B–1) as representative values for typical grades of plutonium that might be used to fabricate MOX fresh fuel. Pure plutonium-239 has been included to contrast the effect of the other plutonium isotopes. Note that in addition to the isotopes identified in Table B–1, plutonium will contain plutonium-236 and americium-241 (from plutonium-241 decay).

Initially, it is expected that MOX fresh fuel will be fabricated using weapons grade (WG) plutonium. A more mature MOX fuel program might be expected to fabricate MOX fresh fuel from previously irradiated WG MOX fuel that may have a composition similar to fuel grade (FG) plutonium. Fabricating MOX fresh fuel from power grade (PG) plutonium would require a much more mature MOX fuel program.

To compare MOX fresh fuel with LEU fresh fuel, we need to choose representative compositions for each fuel type. For a reference LEU fresh fuel, we choose uranium dioxide (UO_2) with 4 weight percent (wt%) U-235 and 96 wt% U-238. For the various grades of plutonium in MOX fresh fuel, we choose UO_2-PuO_2 having 4 wt% Pu-239 with the remaining plutonium isotopes scaled as required by Table B–1, and depleted uranium with 0.2 wt% U-235 and 99.8 wt% U-238. The actual composition of MOX RAM found in practice will not match these compositions, but they are appropriate for comparing the effects of MOX RAM using various grades of plutonium. Table B–2 lists the weight percents for heavy metal isotopes used in this study.

The nuclide depletion and decay code ORIGEN-ARP (Bowman and Leal 2000) can be used to determine the heat generation rates for arbitrary compositions of plutonium with depleted uranium in MOX fresh fuel. Table B–3 lists the ratio of heat generation rates for MOX fresh fuel relative to LEU fresh fuel using the composition weight percents for MOX fresh fuel fabricated from the various plutonium grades from Table B–2 in ORIGEN-ARP. These are the values predicted at the initial time of MOX fuel fabrication when the composition weight percents for the various plutonium isotopes are as given in Table B–2. After these nuclides begin to decay, the heat generation rate decreases with time, so the initial heat generation rate is also the maximum rate.

Table B–1	Typical isotopic mix in weight percent for various grades of plutonium as specified in the 3013 Standard (DOE 2012)			
Isotope	**Pure ^{239}Pu**	**Weapons grade**	**Fuel grade**	**Power grade**
^{238}Pu	0	0.05	0.1	1.0
^{239}Pu	100	93.50	86.1	62.0 [a]
^{240}Pu	0	6.00	12.0	22.0
^{241}Pu	0	0.40	1.6	12.0
^{242}Pu	0	0.05	0.2	3.0

[a]63% reduced to 62% so that the sum is 100%. Source: DOE 2012.

Table B–2	Weight percents for heavy metal isotopes chosen for comparing MOX with LEU for various grades of plutonium				
Nuclide	**No plutonium [a]**	**Pure ^{239}Pu**	**Weapons grade [b]**	**Fuel grade [b]**	**Power grade [b]**
^{235}U	4.0000	0.1920	0.1914	0.1907	0.1871
^{238}U	96.0000	95.8080	95.5305	95.1653	93.3613
^{238}Pu	0.0000	0.0000	0.0021	0.0047	0.0645
^{239}Pu	0.0000	4.0000	4.0000	4.0000	4.0000
^{240}Pu	0.0000	0.0000	0.2567	0.5575	1.4194
^{241}Pu	0.0000	0.0000	0.0171	0.0743	0.7742
^{242}Pu	0.0000	0.0000	0.0021	0.0093	0.1935

[a]No plutonium means LEU oxide with 4 wt% uranium-235 and 96 wt% uranium-238. Note that fresh LEU fuel will normally contain traces of uranium-232, uranium-233, uranium-234, and uranium-236 from recycled and natural uranium. The quantities of these isotopes normally present in fresh LEU are not significant for the comparisons in this appendix.
[b]The plutonium mixtures will also contain plutonium-236 and americium-241. These isotopes can have a significant effect on neutron and/or gamma generation rates.

Table B–3	Ratio of heat generation rate for MOX fresh fuel composed of various grades of plutonium relative to LEU fresh fuel				
Decay time	**No plutonium**	**Pure ^{239}Pu**	**Weapons grade**	**Fuel grade**	**Power grade**
Initial	1	7,300	10,200	13,700	53,900
Maximum	1	7,300	10,200	13,700	53,900

The heat generation rate for any MOX fresh fuel is about four orders of magnitude, or more, greater than that from LEU fresh fuel. Using FG plutonium instead of WG plutonium causes the heat generation rate to increase by about another factor of 1.3. Using PG plutonium instead of FG plutonium causes the heat generation rate to increase by about another factor of 3.9. For reference, 1 metric ton of heavy metal of MOX fuel fabricated from WG plutonium will generate more than 100 watts of decay heat.

The heat is generated predominately by alpha decay of the heavy nuclides. The average alpha energy spectrum for the plutonium isotopes is greater than that for the uranium isotopes by about 25 percent. However, the primary reason heat generation is greater for plutonium is that its specific activity for alpha decay is four to five orders of magnitude larger than that for uranium. Table B–4 shows some specific decay parameters for MOX-relevant nuclides.

The gamma emission code GAMGEN (Gosnell 1990) can be used to determine the gamma emission rates for equal weights of various nuclides of uranium, plutonium, and americium. Shielding for LEU is not a significant problem as a function of decay time. Therefore, studying the gamma emission rate for each nuclide of interest relative to LEU gives a measure of how much more shielding may be required to adequately reduce radiation levels when that nuclide is present than for LEU. Table B–5 lists the gamma emission rates at 20 years of decay time for equal weights of each nuclide, relative to the LEU gamma emission rate at 20 years of decay time, for four energy ranges corresponding to different minimum gamma energies. Although gamma emission rates are not necessarily maximized at 20 years decay time, this decay time was chosen because it gives a better indication of the relation of the various nuclide emission rates relative to LEU with time. The maximum gamma energies for each nuclide are below 3.3 mega electron volts (MeV), and sometimes significantly below. The reason the gamma emission ratios are listed for several different energy ranges is to provide some indication of the energy distribution for the gammas of each nuclide as the minimum gamma energy increases, since more effective or greater amounts of shielding are required as gamma energy increases. This is facilitated by listing the average gamma energy for each nuclide for each energy range in the table. Each nuclide has a different average gamma energy for a given energy range because each has a unique gamma energy spectrum. When the average energy for a nuclide is close to the minimum energy for an energy range, this indicates that most gammas in that range have energies near to that of the minimum energy.

The nuclides plutonium-236 and uranium-232 have very large emission ratios because of the relatively short half-lives and 2.614 MeV gammas emitted after chain decaying to thallium-208. These gammas may require additional package shielding and can usually be tolerated at amounts no greater than about 10^{-4} weight percent of heavy metal nuclides. The nuclides uranium-236, americium-241, uranium-234, and neptunium-237 result from radioactive decay of plutonium-240, plutonium-241, plutonium-238, and americium-241, respectively. The nuclide uranium-233 is usually present in trace quantities.

In Table B–5 for the minimum gamma energies corresponding to 0.041 and 0.183 MeV, most of the nuclides have emission ratios greater than 1.00. The nuclides plutonium-238, plutonium-240, plutonium-241, plutonium-242, uranium-234, uranium-236, and americium-241 have a majority of gammas in the energy range between roughly 0.04 and 0.12 MeV, because their average energies for the first energy range are close to the minimum energy of 0.041 MeV. However, except for uranium-235, all nuclides have average energies greater than 0.28 MeV for the second energy range. Therefore, these nuclides have considerable gammas with energies that will require specific gamma shielding if present in sufficient quantities. This is reinforced by the emission ratios and average energies for the energy range with a minimum gamma energy of 0.498 MeV, particularly for the plutonium isotopes. For the energy range with a minimum gamma energy of 1.000 MeV, only plutonium-236 (except for trace nuclides) has high emission rates of very-high-energy gammas that may require substantial shielding if it is present in a significant quantity.

ORIGEN-ARP also gives the gamma emission rates for arbitrary compositions of plutonium with depleted uranium in MOX fresh fuel. Table B–6 lists the ratio of gamma emission rates for MOX fresh fuel relative to LEU fresh fuel using the composition weight percents for MOX fresh fuel fabricated from the various plutonium grades from Table 2 in ORIGEN-ARP. Table B–4b lists both rates for initial time and maximum rates after some decay time. The decay time at maximum gamma emission rates depends on the plutonium grade in question.

Table B–4   Specific decay parameters for MOX-relevant nuclides

Radionuclide	Half-life (years)	Decay energy (MeV/event)	Decay energy (watt-yr/mole)	Specific heat generation rate (watts/kg)
^{233}U	1.60E+05	4.909	15,021	5.81E-01
^{235}U	7.10E+08	4.681	14,333	6.00E-05
^{238}U	4.50E+09	4.195	12,836	8.00E-06
^{238}Pu	8.78E+01	5.593	17,113	5.67E+02
^{239}Pu	2.41E+04	5.244	16,046	1.93E+00
^{240}Pu	6.54E+03	5.255	16,079	7.10E+00
^{241}Pu	1.44E+01	0.0205	62.7	1.25E+01
^{242}Pu	3.76E+05	4.983	15,246	1.16E-01
^{241}Am	4.32E+02	5.637	17,248	1.15E+02

Table B–5   Gamma emission rates relative to the LEU gamma emission rate and average gamma energies for equal weights of some nuclides of uranium, plutonium, neptunium, and americium at 20 years of decay time

Select nuclides	Gamma energies ≥0.041 MeV		Gamma energies ≥0.183 MeV		Gamma energies ≥0.498 MeV		Gamma energies ≥1.000 MeV	
	Emission relative to LEU	Average energy (MeV)	Emission relative to LEU	Average energy (MeV)	Emission relative to LEU	Average energy (MeV)	Emission relative to LEU	Average energy (MeV)
^{236}Pu	2.73E+08	0.7929	4.46E+08	0.9927	2.90E+09	1.4300	2.26E+09	2.5212
^{238}Pu	5.33E+04	0.0624	1.37E+02	0.7497	1.31E+03	0.7906	7.71E+01	1.1753
^{239}Pu	2.89E+02	0.1173	7.08E+01	0.3883	8.39E+00	0.6968	1.06E-02	1.1750
^{240}Pu	9.05E+02	0.0600	1.66E+00	0.3941	5.97E+00	0.6831	4.05E-09	2.1790
^{241}Pu	5.71E+06	0.0544	4.74E+03	0.2805	2.64E+03	0.6771	8.49E-07	1.4577
^{242}Pu	1.32E+01	0.0613	3.94E-06	0.9783	3.75E-05	1.0389	3.75E-05	1.2507
^{232}U	2.73E+08	0.7930	4.46E+08	0.9927	2.91E+09	1.4300	2.26E+09	2.5212
^{233}U	2.62E+02	0.1801	1.88E+02	0.3549	8.32E+01	1.3620	1.30E+02	1.4607
^{234}U	7.58E+01	0.0789	2.21E-01	0.7588	1.39E+00	1.0282	1.05E+00	1.5553
^{235}U	1.75E+01	0.1901	2.24E+01	0.2402	7.27E-03	0.7422	1.49E-04	1.1750
^{236}U	4.76E-01	0.0723	1.52E-06	0.8876	1.05E-05	1.1865	5.49E-06	2.2074
^{238}U	3.12E-01	0.2289	1.09E-01	0.9783	1.04E+00	1.0389	1.04E+00	1.2507
LEU	1.00E+00	0.2017	1.00E+00	0.3177	1.00E+00	1.0388	1.00E+00	1.2507
^{237}Np	6.06E+03	0.2096	5.94E+03	0.3374	2.63E-04	1.3575	4.09E-04	1.4597
^{241}Am	9.09E+06	0.0543	1.85E+03	0.3905	4.21E+03	0.6771	4.23E-06	1.4586

The gamma emission rates include only gammas with energies equal to or greater than 100 kilo electron volts (keV). The assumption is that gammas with energies less than 100 keV will be absorbed by the normal packaging materials required to transport MOX fresh fuel contents, specifically the strong 59.5 keV gammas coming from any americium-241 produced through decay of plutonium-241. Note that MOX containing plutonium-236 at concentrations greater than about 10^{-4} wt% of total plutonium mass or significant americium-241 ingrowth may have larger gamma emission rates than are shown in Table B–6.

Table B–6	Ratio of gamma emission rate for gamma energies exceeding 100 keV for MOX fresh fuel composed of various grades of plutonium relative to LEU fresh fuel				
Decay time	No plutonium	Pure ^{239}Pu	Weapons grade	Fuel grade	Power grade
Initial	1.0	6.1	6.1	6.9	15.4
Maximum	1.0	6.1	6.1	7.2	83.5

The gamma emission rates for MOX fresh fuel from both WG and FG plutonium are less than an order of magnitude greater than those for LEU fresh fuel. The gamma emission rates for MOX fresh fuel from PG plutonium can be up to about two orders of magnitude greater than those for LEU fresh fuel, depending on the time since MOX fuel fabrication.

The neutron-emission code SOURCES (Wilson et al. 1999) can be used to determine the neutron-emission rates for spontaneous fission and alpha-induced neutrons for equal weights of various nuclides of uranium, plutonium, and americium. Table B–7 lists the neutron-emission rates for spontaneous fission and alpha-induced neutrons from oxygen-17 and oxygen-18, for equal weights of nuclides at the initial MOX fuel fabrication time relative to the LEU neutron-emission rate. Also listed in the table is the average neutron energy for each nuclide and each neutron-emission process.

On an equal weight basis, plutonium-238, plutonium-240, plutonium-242, and americium-241 are overwhelmingly the largest source for neutron emission for the nuclides listed in Table B–7. Most nuclides listed in the table have neutron-emission rates greater than LEU by one or more orders of magnitude. Table B–7 also shows that neutron emissions from uranium isotopes are insignificant relative to those from plutonium isotopes on an equal weight basis. The average neutron energies listed in Table B–7 are between about 1.7 MeV and 2.5 MeV. This means that the spectral energy distribution for neutrons plays a much smaller role than does the spectral energy distribution for gammas.

The neutron-emission code SOURCES can also be used to determine the neutron-emission rates for spontaneous fission and alpha-induced neutrons for arbitrary compositions of plutonium isotopes with depleted uranium in MOX fresh fuel. Table B–8 lists the ratio of neutron-emission rates for MOX fresh fuel relative to LEU fresh fuel using the composition weight percents for MOX fresh fuel fabricated from the various plutonium grades from Table B–2 in SOURCES. Note that MOX fresh fuel with significant americium-241 ingrowth (from plutonium-241 decay) can have significantly larger relative neutron-emission rates, as is shown in the last line of Table B–8.

Replacing 4 wt% uranium-235 with 4 wt% plutonium-239 increases the neutron-emission rate by a factor of about 24. Using WG plutonium instead of pure plutonium-239 causes the neutron-emission rate to increase by about another order of magnitude. Using FG plutonium instead of WG plutonium causes the neutron-emission rate to increase by about another factor of 2. Using PG plutonium instead of FG plutonium causes the neutron-emission rate to increase by about another factor of 3.

Plutonium-241 decays to americium-241 with a half-life of 14.35 years. Americium-241 is a stronger neutron source, so to get a bounding value for the expected increase in neutron-emission rate when plutonium-241 decays to americium-241, all plutonium-241 is replaced with

Table B–7 Neutron-emission rates relative to the LEU neutron-emission rate and average gamma energies for equal weights of some nuclides of uranium, plutonium, and americium for (α, n) with oxygen-17 and oxygen-18, spontaneous fission (SF), and the sum of all three (total) neutron-emission processes

Select nuclides	^{17}O (α, n) relative to LEU	Average energy (MeV)	^{18}O (α, n) relative to LEU	Average energy (MeV)	SF relative to LEU	Average energy (MeV)	Total relative to LEU
^{238}Pu	1.03E+08	2.52	1.27E+08	2.37	1.98E+05	2.02	1.24E+06
^{239}Pu	2.86E+05	2.44	3.62E+05	2.25	1.67E+00	2.07	2.97E+03
^{240}Pu	1.06E+06	2.44	1.33E+06	2.25	7.84E+04	1.93	8.87E+04
^{241}Pu	9.42E+03	2.39	1.23E+04	2.19	3.77E+00	2.00	1.04E+02
^{242}Pu	1.49E+04	2.38	1.94E+04	2.19	1.31E+05	1.96	1.30E+05
^{233}U	3.50E+04	2.37	4.53E+04	2.17	6.23E-02	2.02	3.72E+02
^{235}U	5.96E+00	2.27	6.67E+00	2.07	2.29E-02	1.89	7.80E-02
^{236}U	1.87E+02	2.29	2.23E+02	2.09	4.19E-01	1.83	2.26E+00
^{238}U	7.93E-01	2.20	7.64E-01	1.97	1.04E+00	1.69	1.04E+00
LEU	1.00E+00	2.22	1.00E+00	2.00	1.00E+00	1.74	1.00E+00
^{241}Am	2.05E+07	2.51	2.53E+07	2.36	9.03E+01	2.15	2.09E+05

Table B–8 Ratio of neutron-emission rate for MOX fresh fuel composed of various grades of plutonium relative to LEU fresh fuel

Nuclide composition	No plutonium	Pure ^{239}Pu	Weapons grade	Fuel grade	Power grade
Fresh fuel	1	24	243	506	1,686
^{241}Pu replaced by ^{241}Am	1	24	250	536	1,995

americium-241, and the neutron-emission rate is recalculated for each of these new artificial grades of plutonium.[1] The last row of Table B–8 lists the values obtained. This approach gives an indication of what decay time can do to neutron-emission rates. The effect on neutron-emission rate of plutonium-241 decay to americium-241 is expected to be rather small, except for MOX fresh fuel fabricated from PG plutonium, where it could increase by a factor of about 20 percent.

The uncertainties in the rates of heat generation, gamma emission, or neutron emission from analyses performed using radiation transport codes and cross section sets, such as those employed above, for MOX RAM packages should be comparable to those performed for packages containing LEU RAM for the purposes required for thermal and shielding reviews.

In summary, heat generation and neutron-emission rates increase significantly when MOX RAM replaces LEU RAM. The alpha-energy spectrum responsible for most heat generation is somewhat different for MOX RAM and LEU RAM, but that is not significant in relation to the difference in the magnitude of the heat-generation rates between them. The neutron energy spectra from MOX RAM and LEU RAM are also somewhat different, but, again, this is not

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[1]   Replacing plutonium-241 with americium-241 is bounding for a neutron shielding evaluation but not for a criticality evaluation

significant in relation to the difference in the magnitude of the neutron-emission rates between them. The gamma-emission rate increases between MOX RAM and LEU RAM are not as important so long as plutonium-236 is less than about 10^{-4} wt% of the heavy metal present in MOX RAM. Otherwise, the strong 2.614 MeV gamma from the chain decay of plutonium-236 to thallium-208 becomes an important source of gamma radiation that requires additional package shielding to shield against. However, the gamma energy spectra from MOX RAM and LEU RAM can be quite different depending on the nuclides present, and this can be significant from a shielding point of view.

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