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Storage of Assemblies Containing Mixed-Oxide Fuel

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MANAGED AND OPERATED BY
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STORAGE OF ASSEMBLIES CONTAINING MIXED-OXIDE FUEL

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PREFACE

This document is one in a series of topical reports written in support of the U.S. Department of Energy's (DOE's) *Program Acquisition Strategy for Obtaining Mixed-Oxide Fuel Fabrication and Reactor Irradiation Services* (PAS) [formerly *Procurement Implementation Plan for Acquisition of Mixed-Oxide Fuel Fabrication Services and Reactor Irradiation Services* (PIP)]. This series of topical reports is intended to increase access to available information for parties interested in responding to PAS and the subsequent request for proposal. These topical reports address subjects relevant to DOE's strategy concerning disposition of surplus plutonium by irradiating mixed-oxide (MOX) fuel in existing, domestic commercial reactors.

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1. INTRODUCTION

Both fresh and spent mixed-oxide (MOX) assemblies exhibit physical characteristics that will necessitate additional measurements and analyses beyond those that are done for low-enriched uranium (LEU) assemblies. While none of these analyses or measurements require technological development, estimates for disposition costs and schedules must account for these factors. This report identifies those nuclear engineering analyses that will be required to demonstrate acceptability of MOX fuel assembly storage.

2. FRESH FUEL STORAGE

The reference fresh fuel storage scheme as documented in Ref. 1 is to construct a stand-alone, ex-reactor fuel storage vault. This vault is deemed necessary by the reactor vendors because of the perceived requirement of increased security associated with MOX fuel. The building is to serve as both temporary storage for fresh fuel and dry storage for spent fuel. If, indeed, the facility is to be designed for dry storage, regulatory requirements are available in Refs. 2 and 3.

Criticality safety calculations will be required for MOX fresh fuel storage. Reference 4 provides guidance on validation of criticality safety codes for application to transportation and storage. Although the decay heat generation in fresh MOX fuel will be significantly greater than for LEU fuel, engineered (forced) cooling systems should not be required. A mechanism for transportation of assemblies between the MOX storage facility and the reactor building must be devised, because none was specified in Ref. 1.

The operators of the Tennessee Valley Authority's Bellefonte plant have expressed the opinion that, at their facility, a separate storage facility will not be required. Even if a separate facility is not required, it is likely that significant modifications to existing facilities would be required. In a recent visit to the Arkansas Nuclear One site, it was noted that in order to receive MOX assemblies transported via a safe, secure trailer (SST), it would be necessary to modify the fuel receiving facility and its administrative controls to provide the appropriate safeguards controls for the material in the receiving bay, thus removing the need to isolate the receiving bay from the refueling floor during fuel handling operations. As such, it would be necessary to fortify the receiving bay to Category I safeguards measures. Such modifications would also include construction of appropriate facilities to handle SST operations at the reactor. Some information regarding safeguards criteria can be found in Ref. 1.

Also for Arkansas Nuclear One, the fuel vault has only the capacity to hold a partial core. If a full core of new fuel is received, some will have to be moved into the spent fuel pool in advance of charging the reactor core. For MOX fuel, the spent fuel pool storage spacing may be insufficient for effective criticality control. This spacing would have to be reviewed prior to any such fuel receipt.

3. SPENT FUEL STORAGE

Regardless of whether the spent fuel is stored in a pool or in dry storage, nuclear engineering evaluations of the storage sites must consider criticality safety, radiation protection (shielding), decay heat generation, and potential radionuclide releases to the environment in the event of an accident. Analyses for each of these four areas will be facility and fuel cycle dependent. However, an estimate of the impact of substituting MOX fuel for LEU fuel has been calculated by J. C. Ryman at Oak Ridge National Laboratory. Based on the General Electric boiling-water reactor (BWR) MOX fuel design described in Ref. 1 (BWR-5) and an LEU fuel cycle designed to yield equivalent burnup (37.6 GWd/MT), comparisons were made between MOX and LEU fuel cycles and are presented in subsequent tables.

3.1 ISOTOPIC COMPOSITION OF SPENT FUEL

Discharge spent fuel inventories for MOX and LEU fuels are shown in Table 1. Because an LEU assembly is charged with much more ^{235}U, it burns more ^{235}U; it will, therefore, contain more ^{236}U and

Table 1. Nuclides of interest for criticality safety

Nuclide	Quantity of discharge per assembly (g)		Ratio of MOX to LEU
	MOX	LEU	
^{16}O	4.72×10^{4}	4.84×10^{4}	0.975
^{95}Mo	1.25×10^{2}	1.53×10^{2}	0.817
^{101}Ru	1.63×10^{2}	1.60×10^{2}	1.02
^{99}Tc	1.52×10^{2}	1.59×10^{2}	0.956
^{103}Rh	1.23×10^{2}	8.82×10^{1}	1.39
^{109}Ag	3.71×10^{1}	1.90×10^{1}	1.95
^{131}Xe	9.42×10^{1}	8.74×10^{1}	1.08
^{133}Cs	2.24×10^{2}	2.30×10^{2}	0.974
^{143}Nd	1.38×10^{2}	1.57×10^{2}	0.879
^{145}Nd	1.16×10^{2}	1.37×10^{2}	0.847
^{147}Sm	2.08×10^{1}	2.24×10^{1}	0.929
^{149}Sm	0.444	0.398	1.12
^{150}Sm	6.17×10^{1}	6.12×10^{1}	1.01
^{151}Sm	3.39	2.83	1.20
^{152}Sm	3.11×10^{1}	2.61×10^{1}	1.19
^{151}Eu	6.90×10^{-3}	5.02×10^{-3}	1.37
^{153}Eu	2.87×10^{1}	2.27×10^{1}	1.26
^{154}Eu	1.15×10^{1}	8.72	1.32
^{155}Gd	0.139	0.0330	4.21
^{233}U	3.64×10^{-5}	5.01×10^{-4}	0.0727
^{234}U	0.457	2.87×10^{1}	0.0159
^{235}U	1.04×10^{2}	1.31×10^{3}	0.0794
^{236}U	4.39×10^{1}	8.16×10^{2}	0.0538
^{238}U	1.68×10^{5}	1.72×10^{5}	0.977
^{237}Np	3.08×10^{1}	9.93×10^{1}	0.310
^{238}Pu	3.66×10^{1}	4.23×10^{1}	0.865
^{239}Pu	1.40×10^{3}	9.81×10^{2}	1.43
^{240}Pu	1.16×10^{3}	4.49×10^{2}	2.58
^{241}Pu	5.57×10^{2}	2.64×10^{2}	2.11
^{242}Pu	2.43×10^{2}	1.11×10^{2}	2.19
^{241}Am	4.20×10^{1}	1.49×10^{1}	2.82
^{242m}Am	1.17	0.414	2.83
^{243}Am	5.97×10^{1}	2.68×10^{1}	2.23
^{245}Cm	0.974	0.349×10^{-3}	2.79

^{237}Np at discharge than a MOX assembly. At long decay times, the amounts of ^{235}U, ^{236}U, and ^{237}Np increase significantly in the MOX assembly as compared with the LEU assembly because of the alpha decay of ^{239}Pu, ^{240}Pu, and ^{241}Am (which arises from the decay of ^{241}Pu); the ^{237}Np becomes absolutely greater in the MOX assembly.

The MOX assembly has about 1.8 times as much plutonium at discharge as the LEU assembly. All of the americium and curium isotopes show higher concentrations at discharge in the MOX fuel relative to the LEU fuel.

3.2 ASSEMBLY ACTIVITY

Table 2 gives discharge activities for nuclides of interest in both MOX and LEU assemblies. Most of the contribution to activity comes from actinides and fission products, with light elements contributing 1% or less.

Table 2. Discharge activities for BWR-5 MOX and LEU assemblies

Nuclide	Activity in MOX per assembly (Ci)	Activity in LEU per assembly (Ci)	Ratio of MOX to LEU
^{3}H	1.01×10^{2}	9.41×10^{1}	1.07
^{14}C	3.17×10^{-2}	4.07×10^{-2}	7.79×10^{-1}
^{58}Co	3.94×10^{2}	3.83×10^{2}	1.03
^{60}Co	5.51×10^{2}	5.49×10^{2}	1.00
^{85}Kr	9.77×10^{2}	1.65×10^{3}	5.92×10^{-1}
^{85m}Kr	1.86×10^{4}	2.74×10^{4}	6.79×10^{-1}
^{87}Kr	3.63×10^{4}	5.44×10^{4}	6.67×10^{-1}
^{88}Kr	4.85×10^{4}	7.49×10^{4}	6.48×10^{-1}
^{86}Rb	1.39×10^{2}	2.42×10^{2}	5.74×10^{-1}
^{89}Sr	5.26×10^{4}	8.51×10^{4}	6.18×10^{-1}
^{90}Sr	7.55×10^{3}	1.48×10^{4}	5.10×10^{-1}
^{91}Sr	9.13×10^{4}	1.33×10^{5}	6.86×10^{-1}
^{92}Sr	1.08×10^{5}	1.45×10^{5}	7.45×10^{-1}
^{90}Y	8.53×10^{3}	1.61×10^{4}	5.30×10^{-1}
^{91}Y	7.40×10^{4}	1.09×10^{5}	6.79×10^{-1}
^{92}Y	1.08×10^{5}	1.46×10^{5}	7.40×10^{-1}
^{93}Y	9.07×10^{4}	1.14×10^{5}	7.96×10^{-1}
^{95}Zr	1.47×10^{5}	1.70×10^{5}	8.65×10^{-1}
^{97}Zr	2.04×10^{5}	2.20×10^{5}	9.27×10^{-1}
^{95}Nb	1.22×10^{5}	1.41×10^{5}	8.65×10^{-1}
^{99}Mo	2.22×10^{5}	2.31×10^{5}	9.61×10^{-1}
^{99m}Tc	1.95×10^{5}	2.04×10^{5}	9.56×10^{-1}
^{103}Ru	2.11×10^{5}	1.87×10^{5}	1.13
^{105}Ru	1.93×10^{5}	1.53×10^{5}	1.26
^{106}Ru	1.15×10^{5}	7.52×10^{4}	1.53
^{105}Rh	1.86×10^{5}	1.46×10^{5}	1.27
^{127}Sb	1.35×10^{4}	1.18×10^{4}	1.14
^{127}Te	1.28×10^{4}	1.11×10^{4}	1.15
^{127m}Te	1.74×10^{3}	1.46×10^{3}	1.19
^{129}Te	4.08×10^{4}	3.90×10^{4}	1.05
^{129m}Te	7.74×10^{3}	7.32×10^{3}	1.06
^{132}Te	1.76×10^{5}	1.79×10^{5}	9.83×10^{-1}
^{131}I	1.24×10^{5}	1.25×10^{5}	9.92×10^{-1}
^{132}I	1.80×10^{5}	1.83×10^{5}	9.84×10^{-1}
^{133}I	2.45×10^{5}	2.55×10^{5}	9.61×10^{-1}
^{134}I	2.63×10^{5}	2.79×10^{5}	9.43×10^{-1}
^{135}I	2.37×10^{5}	2.43×10^{5}	9.75×10^{-1}
^{131m}Xe	1.40×10^{3}	1.40×10^{3}	1.00
^{133m}Xe	7.99×10^{3}	8.14×10^{3}	9.82×10^{-1}
^{133}Xe	2.36×10^{5}	2.45×10^{5}	9.63×10^{-1}
^{135}Xe	8.25×10^{4}	7.61×10^{4}	1.08
^{134}Cs	2.65×10^{4}	2.74×10^{4}	9.67×10^{-1}
^{136}Cs	1.10×10^{4}	9.06×10^{3}	1.21

Table 2. (continued)

Nuclide	Activity in MOX per assembly (Ci)	Activity in LEU per assembly (Ci)	Ratio of MOX to LEU
^{137}Cs	2.14×10^{4}	2.17×10^{4}	9.86×10^{-1}
^{139}Ba	2.05×10^{5}	2.21×10^{5}	9.28×10^{-1}
^{140}Ba	2.08×10^{5}	2.21×10^{5}	9.41×10^{-1}
^{140}La	2.21×10^{5}	2.36×10^{5}	9.36×10^{-1}
^{141}La	1.86×10^{5}	2.01×10^{5}	9.25×10^{-1}
^{142}La	1.78×10^{5}	1.96×10^{5}	9.08×10^{-1}
^{141}Ce	1.72×10^{5}	1.86×10^{5}	9.25×10^{-1}
^{143}Ce	1.63×10^{5}	1.85×10^{5}	8.81×10^{-1}
^{144}Ce	1.04×10^{5}	1.24×10^{5}	8.39×10^{-1}
^{143}Pr	1.57×10^{5}	1.78×10^{5}	8.82×10^{-1}
^{147}Nd	7.85×10^{4}	8.26×10^{4}	9.50×10^{-1}
^{237}U	2.97×10^{4}	1.18×10^{5}	2.52×10^{-1}
^{239}U	2.61×10^{6}	2.74×10^{6}	9.53×10^{-1}
^{237}Np	2.17×10^{-2}	7.00×10^{-2}	3.10×10^{-1}
^{238}Np	1.75×10^{4}	6.03×10^{4}	2.90×10^{-1}
^{239}Np	2.60×10^{6}	2.74×10^{6}	9.49×10^{-1}
^{238}Pu	6.28×10^{2}	7.25×10^{2}	8.66×10^{-1}
^{239}Pu	8.72×10^{1}	6.09×10^{1}	1.43
^{240}Pu	2.63×10^{2}	1.02×10^{2}	2.58
^{239}U	2.61×10^{6}	2.74×10^{6}	9.53×10^{-1}
^{237}Np	2.17×10^{-2}	7.00×10^{-2}	3.10×10^{-1}
^{238}Np	1.75×10^{4}	6.03×10^{4}	2.90×10^{-1}
^{239}Np	2.60×10^{6}	2.74×10^{6}	9.49×10^{-1}
^{238}Pu	6.28×10^{2}	7.25×10^{2}	8.66×10^{-1}
^{239}Pu	8.72×10^{1}	6.09×10^{1}	1.43
^{240}Pu	2.63×10^{2}	1.02×10^{2}	2.58
^{241}Pu	5.76×10^{4}	2.73×10^{4}	2.11
^{241}Am	1.44×10^{2}	5.11×10^{1}	2.82
^{242}Cm	3.22×10^{4}	1.25×10^{4}	2.58
^{244}Cm	1.63×10^{3}	6.69×10^{2}	2.44

The activity from structural materials is comparable for both assembly types. If everything else were equal, the light-element contribution to activity would just be proportional to burnup. The amount of light elements per metric ton of heavy metal (MTHM) is the same for both assemblies, and the burnup is the same. However, there is 0.183 MTHM in the LEU assembly and 0.176 MTHM in the MOX assembly, which causes slightly more light-element activity in the LEU assembly. A major contributor to light-element activity is ^{60}Co. The final amount of ^{60}Co depends on the initial amount of ^{59}Co in the assembly.

Activities for many, but not all, fission products are comparable, with the LEU assembly having somewhat higher values (4 to 5%) than the MOX assembly. These ratios of fission product activity (MOX to LEU) are just about the same as the ratio of heavy metal content (0.176 MTHM to 0.183 MTHM = 0.962) for the MOX and LEU assemblies because the burnups are equal. The slight differences from the

value of 0.962 are caused by differences in the fission yields of the uranium and plutonium mixtures in the two assemblies.

The actinide activity is slightly greater at discharge (4%) for the LEU assembly because of higher activities for the short-lived nuclide ^{239}Np and its daughter ^{239}U. For decay times from 1 year until 10,000 years, the larger quantities of long-lived actinides in the MOX assembly cause its actinide activity to be greater by about a factor of 2. Beyond that decay time, the ratio of MOX to LEU actinide activity slowly decreases to a value of about 1.1 at 250,000 years.

3.3 DECAY HEAT

The actinide decay heats for MOX and LEU assemblies are comparable at discharge, but are consistently larger for the MOX assembly at long decay times. The fission product decay heat is generally less in the MOX assembly for decay times less than 300 years and greater past that time. The total decay heat at discharge is about 6% larger for the LEU assembly because it comes primarily from fission products at that time. For cooling times of 1 year or greater, the total is greater for the MOX assembly and is comprised principally of actinide decay heat for times greater than 100 years.

3.4 DOSE RATES

The neutron dose rate from the MOX assembly is about 2.5 times greater than that from the LEU assembly. The greater dose from the MOX assembly is caused by the greater amounts of curium and americium nuclides in that fuel as compared with the LEU assembly. The gamma dose rates from both assemblies are comparable (ratios from 1.07 to 0.95) from discharge until 300 years following discharge (see Table 3). Neutron dose rates are 1/100,000th of the gamma dose rate at discharge but are 1/200th of the gamma dose rate at 300 years after discharge.

Table 3. Gamma dose rate 1 m from a bare BWR-5 assembly at the axial midplane

	Dose rate (rem h^{-1})	
Time after discharge	MOX	LEU
At discharge	1.55×10^6	1.66×10^6
1 day	1.39×10^5	1.43×10^5
1 year	5280	4890
5 years	1680	1610
10 years	968	936
100 years	85.1	85.6
300 years	0.927	0.882

3.5 POTENTIAL INHALATION AND INGESTION HAZARDS

Calculations performed using the ORIGEN-S program[5] give a rough estimate of the potential hazards from inhalation and ingestion of radionuclides in the fuel assemblies. These potential hazards are quantified as the amount of air or water necessary to dilute the amount of radionuclides to the radioactivity concentration guide limits of the pre-1993 version of Title 10 of the *Code of Federal Regulations* Part 20. It must be emphasized that these are potential hazards because thorough pathway and dosimetry analyses would be necessary to estimate the doses and risks for particular exposure incidents. Broadly speaking, the MOX assembly is potentially more hazardous for inhalation because of the greater quantities of alpha-emitting

and bone-seeking actinides. The potential ingestion hazard is slightly greater for the nuclides from the LEU assembly because of the importance of light-element and fission product nuclides.

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