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FUEL CYCLE POTENTIAL WASTE FOR DISPOSITION

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Abstract

The United States (U.S.) currently utilizes a once-through fuel cycle where used nuclear fuel (UNF) is stored on-site in either wet pools or in dry storage systems with ultimate disposal in a deep mined geologic repository envisioned. Within the Department of Energy's (DOE) Office of Nuclear Energy (DOE-NE), the Fuel Cycle Research and Development Program (FCR&D) develops options to the current commercial fuel cycle management strategy to enable the safe, secure, economic, and sustainable expansion of nuclear energy while minimizing proliferation risks by conducting research and development of advanced fuel cycles, including modified open and closed cycles.

The safe management and disposition of used nuclear fuel and/or nuclear waste is a fundamental aspect of any nuclear fuel cycle. Yet, the routine disposal of used nuclear fuel and radioactive waste remains problematic. Advanced fuel cycles will generate different quantities and forms of waste than the current LWR fleet.

This study analyzes the quantities and characteristics of potential waste forms including differing waste matrices, as a function of a variety of potential fuel cycle alternatives including:

- Commercial UNF generated by uranium fuel light water reactors (LWR). Four once through fuel cycles analyzed in this study differ by varying the assumed expansion/contraction of nuclear power in the U.S.
- Four alternative LWR used fuel recycling processes analyzed differ in the reprocessing method (aqueous vs. electro-chemical), complexity (Pu only or full transuranic (TRU) recovery) and waste forms generated.
- Used Mixed Oxide (MOX) fuel derived from the recovered Pu utilizing a single reactor pass.
- Potential waste forms generated by the reprocessing of fuels derived from recovered TRU utilizing multiple reactor passes.

Introduction

The safe management and disposition of used nuclear fuel and/or nuclear waste is a fundamental aspect of any nuclear fuel cycle; however, the routine disposal of used nuclear fuel and radioactive waste remains problematic. Advanced fuel cycles will generate different quantities and forms of waste than the current light water reactor (LWR) fleet. This study analyzes the quantities and characteristics of potential waste forms including differing waste matrices, as a function of a variety of potential fuel cycles. The information presented in this paper is more completely investigated in references 1 and 2.

Commercial Light Water Reactor Used Nuclear Fuel – Once-Thru Fuel Cycle

Commercial nuclear power plants have operated in the United States since about 1960. There are currently 104 operating nuclear power plants. Used nuclear fuel (UNF) from these operating plants is currently stored on site in pools or dry storage casks with disposal in a geologic repository envisioned in a once-thru fuel cycle. In addition, UNF from 14 shutdown reactors is currently stored on the reactor sites and at the General Electric wet storage facility at Morris, Il.

Current LWR Uranium Oxide Used Fuel Inventory

The source of current inventory data for this study is information collected in support of the Department of Energy Office of Civilian Radioactive Waste Management's (OCRWM) efforts for licensing the Yucca Mountain Repository [3]. Information collected from RW-859 forms is available on an assembly basis for UNF discharges from 1968 through 2002. Data is also available that was collected to support RW activities on a batch basis for fuel discharges from 1968 through April 2005 [4].

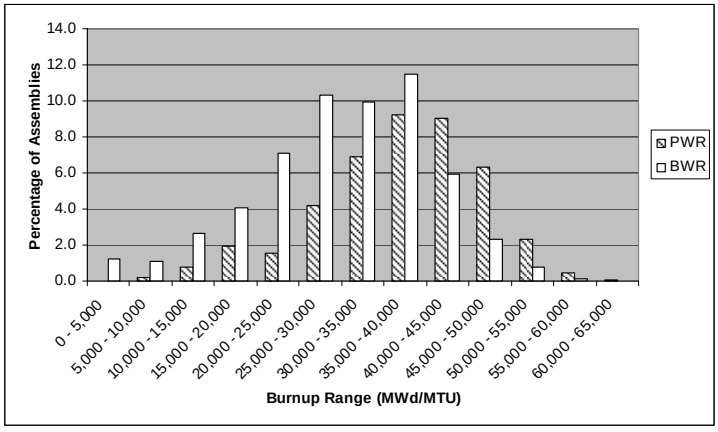
To develop an inventory estimate through 2009, fuel discharge predictions developed for the Nuclear Energy Institute in 2005 were used to estimate the number of assemblies and metric tons of uranium [5]. To estimate the average enrichment and burnup through 2009, projections made by utilities as part of the RW-859 surveys were used. These projections are documented in OCRWM's "Calculation Method for the Projection of Future Spent Fuel Discharges", February 2002 [6]. These projections identified a burnup increase of 2.38% per year for boiling water reactor (BWR) fuel and 1.11% per year for pressurized water reactor (PWR) fuel through 2009. The enrichment increased at the same rate as burnup. Comparison of these projections made in 1998 to actual data collected through 2004 show very good agreement (PWR - actual 46,950 MWd/MTU vs projected 46,922 MWd/MTU; BWR - actual 43,447 MWd/MTU vs 42,787 projected MWd/MTU). Table 1 provides an estimate of the commercial UNF discharged through December 2009. Figure 1 provides a distribution of this estimated inventory as a function of burn-up. Nearly 100% of the fuel currently being discharged exceeds the "high burn-up" threshold of 45,000 MWd/MT.

Table 1 Commercial UNF Estimated Discharge Through 2009

Total Number of Assemblies [a]			**Total Initial Uranium (MTU)** [b]			**Average Enrichment**		**Average Burnup (MWd/MTU)** [c]	
PWR	BWR	Totals	PWR	BWR	Totals	PWR	BWR	PWR	BWR
94,800	124,000	218,800	41,100	22,100	63,200	3.72	3.07	39,400	32,700

a the number of assemblies has been rounded to the nearest 200, totals may not appear to sum correctly
b the estimated fuel discharged has been rounded to the nearest 100 metric tons of uranium (MTU), totals may not appear to sum correctly
c the burn-up has been rounded to the next 100 megawatt-days/MTU (MWd/MTU)

Figure 1 Percentage of Assemblies per Burnup Range in the Current Inventory



Future LWR Uranium Oxide Used Fuel Projections

Future UNF discharge predictions were developed using the same methods used for estimating the current UNF inventory. These projections identified a burnup increase of 2.38% per year for BWR UNF and 1.11% per year for PWR UNF. The enrichment increased at the same rate as burnup until reaching the current enrichment limit of 5%. Once the 5% enrichment limit is reached, the enrichment and burnup are assumed to remain constant.

To provide a range of UNF requiring disposal, four nuclear energy scenarios were selected from those previously evaluated by DOE.

Scenario 1 assumes no replacement of existing nuclear generation reactors. The existing plants are assumed to have one 20 year life extension and will be decommissioned after 60 years of operation. Applying these assumptions the last nuclear generator finishes operations in 2055.

Scenario 2 assumes the amount of current nuclear generation is maintained at the current levels (100 gigawatt-electric/year (GWe/yr)) with new reactors replacing the existing reactors as the existing reactors are decommissioned. The current PWR/BWR ratio is maintained.

Scenario 3 assumes the amount of nuclear generation will increase to 200 GWe/yr from 2020 to 2060.

Scenario 4 assumes the amount of nuclear generation will increase to 400 GWe/yr from 2020 to 2060.

In both scenarios 3 and 4 new nuclear generating capacity is assumed to come on line in 2020 and increase linearly till 2060. Nuclear generation is assumed to remain constant past 2060. Table 2 provides the mass (metric tons of uranium) projected to be generated annually in 2060 (at the assumed completion of nuclear expansion) and cumulatively through the end of the century.

Table 2 Commercial UNF Projections

	Scenario			
	No Replacement	**100 GWe/yr**	**200 GWe/yr**	**400GWe/yr**
Annual Generation Rate at 2060	0	2,300	4,500	9,100
Cumulative UNF (MTU)	140,000	270,000	407,000	682,000

Process Waste Generated by Reprocessing Commercial Light Water Reactor Fuel

To provide a tool for evaluation of the impact of reprocessing, unit quantities of 24 representative LWR UNF compositions have been converted into their equivalent waste forms for a variety of reprocessing methods. For each of the representative fuels the mass, volume, container count and decay heat in each container has been projected for each of the baseline waste forms anticipated.

Reprocessing Methods

Commercial light water reactor UNF reprocessing methods vary in process complexity and technical maturity. Generally the objective of additional complexity is to lessen the potential environmental impact of the resulting waste disposition activities. To support future evaluations of potential environmental impact three aqueous reprocessing methods and one electro-chemical reprocessing method were selected.

Co-Extraction represents the simplest and most technically mature aqueous reprocessing method evaluated. The process envisioned is similar to the current generation of deployed reprocessing technology (e.g., the Rokkasaho Reprocessing Facility). Uranium and plutonium are recovered together (no pure plutonium separation). The principle fission product wastes including the minor actinides are combined with the undissolved solids (UDS) and recovered Tc into a single borosilicate glass wasteform. The gaseous radionuclides I-129 and H-3 released during reprocessing are captured and converted to waste forms suitable for disposal while C-14 and Kr-85 are assumed to be released to the atmosphere.

New Extraction is an advanced aqueous process which recovers all of the transuranic (TRU) elements for re-use. The process envisioned includes Transuranic Extraction (TRUEX) and the Trivalent Actinide Lanthanide Separation by Phosphorus-based Aqueous Komplexes [*sic*.] (TALSPEAK) process for complete TRU recovery. The principle fission product wastes are combined with the UDS and separated Tc into a single borosilicate glass wasteform. The principle gaseous radionuclides I-129, Kr-85, C-14 and H-3 released during reprocessing are captured and converted to waste forms suitable for disposal.

Uranium Extraction (UREX) is an advanced aqueous process which also recovers all of the TRU and in addition separates the fission product waste components into three segments. The Fission Product Extraction (FPEX) process is added to separate the Cs/Sr/Ba/Rb, which is converted to a solid ceramic waste form. The Tc and Undissolved Solids (UDS) are combined with a portion of the zirconium hulls/stainless steel hardware to form a metal alloy, and the remaining fission products are converted to a borosilicate glass. UREX is the most complex of the three aqueous processes evaluated.

Electro-chemical processing is a dry process using conductive molten salt baths to recover all the TRU elements. In this process the fission products are split between three waste streams. Elements which are more noble (as measured by electro-chemical potential) than uranium, such as fuel cladding and noble metal fission products, remain as metals and are incorporated into a metal alloy waste form. Elements less noble than uranium are converted to chloride salts. The lanthanide elements are recovered from the salt by electrolysis and converted to a lanthanide glass. Excess salt is purged; the chloride is adsorbed by zeolite and bonded with glass to make the final waste form. The principle gaseous radionuclides I-129, Kr-85, C-14 and H-3 released during reprocessing are captured and converted to a wasteform suitable for disposal, although most of the I-129 in this process is not released to the gaseous phase but is converted to a molten salt and purged with the excess salt.

Representative Fuels

The historical and projected UNF inventory were reviewed to select fuels representative of the anticipated fuel type, burn-up and age at the time of reprocessing. Three fuel burn-ups were selected to represent the historical and future PWR (20, 40 and 60 GWd/MTU) and BWR (15, 30 and 50 GWd/MTU) reactors discharge. The time of reprocessing is unknown and a broad range potential ages (5, 30, 100 and 500 years) were selected. This broad range of potential burn-ups and ages allows evaluation of various strategic decisions.

Baseline and Alternative Wasteforms

The Global Nuclear Energy Partnership Integrated Waste Management Strategy Baseline Study [7] summarized the state-of-the-art in stabilization concepts for byproduct and waste streams, and recommended a baseline of waste forms for the safe disposition of proposed waste streams from future fuel recycling processes. This baseline has been adopted for this study as applicable to the specific reprocessing method.

The baseline wasteforms include four specific to the principle radionuclides of interest (H-3, C-14, Kr-85 and I-129) released during fuel chopping and other front end processing. There is considerable uncertainty in the need to capture and treat both the C-14 and Kr-85 released during reprocessing with many factors (e.g. reprocessing facility location and environmental

regulations) influencing the final decision. These waste forms provide a comprehensive range of the waste quantities for potential disposal.

The baseline includes two metal wasteforms for separated structural components of the fuel assemblies. These include simple compaction (following decontamination) and a more advanced metal alloy which has been proposed for the UREX process that contains the UDS and Tc. A portion of the hulls and hardware are diverted from the compaction line to provide the zirconium and iron required to produce a durable waste form.

A metal alloy is also used in the electrochemical process. Those elements which are more noble (as measured by electro-chemical potential) than uranium such as the hulls, hardware and noble metal fission products remain as metals. The metal waste is decontaminated by volatilizing adhered salts.

Four fission product waste forms are included. Borosilicate glass is the accepted standard for reprocessing waste disposal; however, the waste loading is limited to avoid the formation of multi-phase glasses. These limits include: 1) a maximum decay heat of 14,000 watts per 2 ft diameter canister to prevent the canister centerline temperature from reaching the transition temperature 2) the molybdenum trioxide solubility is limited to 2.5% by weight, and 3) the noble (Ag, Pd, Rh, Ru) metals are limited to 3% by weight. The limit selected for any representative fuel allows the maximum waste loading and minimum projected waste volume and mass.

UREX processing proposed separating the Cs/Sr/Ba/Rb to segregate the high heat producing waste. The waste is treated by mixing the waste with bentonite clay followed by high pressure pressing and high temperature sintering to produce a ceramic puck.

The electrochemical process utilizes two fission product waste forms. Excess salt is purged with fission products which have been adsorbed onto zeolite. Additional zeolite is added to sequester the excess salt chloride and then bonded with borosilicate glass. The electrochemical process also separates lanthanides which are converted to a lanthanide based glass.

The complete results of this analysis are provided in reference 1. Tables 3 summarizes the borosilicate glass fission product waste form generated by the co-extraction and new extraction reprocessing methods. Trends include:

- Projections of borosilicate glass quantity from the co-extraction process is limited by decay heat (14,000 watts per canister) when “young” UNF is processed.
- Projections of borosilicate glass quantity from the co-extraction processing of UNF older than 30 years are limited by molybdenum trioxide solubility at (2.5 wt %). In these cases the mass, volume and containers per metric ton is a constant regardless of the age of the UNF processed; although, the decay heat continues to decline with fuel age.
- Many of the fission product waste forms included in this study significantly exceed the 1,500 watts/canister limit of the Yucca Mountain license application. Disposal in another undetermined alternative repository could require additional decay storage time prior to disposal or a more dilute waste form.
- Recovery of the Am/Cm in other aqueous reprocessing methods and separation of the Cs/Sr in the UREX reprocessing (results provided in reference 1) reduces the decay heat such that the waste loading is limited by molybdenum trioxide solubility at (2.5 wt %). In these cases the mass, volume and containers per metric ton is a constant regardless of the age of the UNF processed; although, the decay heat continues to decline with fuel age.

Trends in the captured off gas and metal waste forms are discussed in reference 1 along with nine alternative waste forms.

Table 3 Fission Product Waste Trends for Pressurized Water Reactor Used Nuclear Fuel for the Co-Extraction and New Extraction Recycling Methods

		Co- Extraction Borosilicate Glass				New Extraction Borosilicate Glass			
		Containers: 2 ft diameter x 15 ft tall canisters. Each Canister Contains 2,900 kg.				Containers: 2 ft diameter x 15 ft tall canisters. Each Canister Contains 2,900 kg.			
Burn-up (GWd/MTU)	Age (years)	Mass (kg/MT)	Volume (ft^3/MT)	Containers per MT	Decay Heat (W/container)	Mass (kg/MT)	Volume (ft^3/MT)	Containers per MT	Decay Heat (W/container)
20	5	198.53	3.22	0.07	14,000	139.30	2.26	0.05	12,124
	30	147.61	2.40	0.05	7,766	139.30	2.26	0.05	2,911
	100	147.61	2.40	0.05	3,059	139.30	2.26	0.05	562
	500	147.61	2.40	0.05	1,004	139.30	2.26	0.05	0
40	5	410.33	6.66	0.14	14,000	253.53	4.12	0.09	13,111
	30	268.66	4.36	0.09	8,367	253.53	4.12	0.09	3,157
	100	268.66	4.36	0.09	2,928	253.53	4.12	0.09	601
	500	268.66	4.36	0.09	884	253.53	4.12	0.09	0
60	5	658.47	10.69	0.23	14,000	366.12	5.95	0.13	13,851
	30	387.97	6.30	0.13	8,667	366.12	5.95	0.13	3,233
	100	387.97	6.30	0.13	2,546	366.12	5.95	0.13	611
	500	387.98	6.30	0.13	654	366.13	5.95	0.13	0

Mixed Oxide Used Nuclear Fuels

Mixed oxide (MOX) fuel, is a blend of oxides of plutonium and natural uranium, reprocessed uranium, or depleted uranium. This study examines two potential sources of fissile material.

Recycling LWR uranium oxide (UOX) used fuel (burned to 51 GWd/MT and allowed to cool for 5 years post-irradiation) recovers plutonium and uranium which can then be fabricated into a MOX fuel. This MOX fuel is stored for 2 years prior to introduction into the full MOX core. The delay time results in the build-up of Am-241 in the MOX fuel, which arises from the decay of Pu-241. The burnup of the MOX core is limited to 50 GWd/MT because of a constraint on the plutonium content in the MOX fuel. The average plutonium enrichment is 10.74%; therefore, each metric ton of LWR fuel which is reprocessed allows fabrication of 108.9 kg of MOX fuel.

Recognizing the threat of surplus weapons grade plutonium to global security, the governments of the United States of America and the Russian Federation agreed to pursue a long-term disposition initiative limiting the availability of this material for weapons use. The U.S. MOX fuel program will dispose of 34 metric tons (MT) of weapons grade plutonium. To dispose of this quantity of plutonium the program will produce approximately 1,684 fuel assemblies [8]. Each assembly will contain 462.2 kg of heavy metal (HM) per unirradiated fuel assembly [8]. A total of approximately 77.8MT HM of MOX fuel will be generated.

Potential Waste Derived from Reprocessing Advanced Burner Reactor UNF

A key attribute of the “fully closed” nuclear fuel cycle is that no UNF is disposed, only UNF reprocessing wastes are disposed. To investigate reducing the long term (vs. transitional) TRU disposal burden on the repository, this study selected sodium cooled fast reactors with design features and operating parameters such that a burning TRU conversion ratio of 0.5 and 0.75 are achieved. Both oxide and metal fuel forms are used and these fuel types are “associated” with aqueous and electro-chemical reprocessing technologies respectively. While this association is not technically mandated, using this association does allow the differences in the reprocessing methods to be examined.

Tables 4 and 5 summarize the fission product waste forms which may be potentially generated by recycling advanced burner reactor (ABR) fuel. Off gas and metal waste forms are investigated more completely in reference 1.

Table 4 Fission Product Waste Forms from Aqueous Recycling Advanced Burner Reactor UNF

Burn-up (GWd/MTU)	CR	Aqueous Borosilicate Glass Containers: 2 ft diameter x 15 ft tall canisters. Each Canister Contains 2,900 kg. Mass (kg/MT)	Volume (ft^3/MT)	Containers per MT	Decay Heat (W/container)
Oxide Based Fuel					
131	0.75	1,690	27.45	0.58	14,000
166	0.50	2,090	33.95	0.72	14,000

Table 5 Fission Product Waste Forms from Electrochemical Recycling Advanced Burner Reactor UNF

Burn-up (GWd/MTU)	CR	Electrochemical Glass Bonded Zeolite Containers: 2 ft diameter x 15 ft tall canisters. Each Canister Contains 2,900 kg. Mass (kg/MT)	Volume (ft^3/MT)	Containers per MT	Decay Heat (W/container)	Lanthanide Glass Containers: 6in diameter x 60in tall canisters. Each Canister Contains 500 kg. Mass (kg/MT)	Volume (ft^3/MT)	Containers per MT	Decay Heat (W/container)
Metal Based Fuel									
99.6	0.75	2,641	42.77	0.91	2900	58.39	0.46	0.12	21,175
132	0.50	3,368	54.53	1.16	3368	73.14	0.57	0.15	21,574

Future Activities

Efforts will continue in 2011 to evaluate other fuel cycle activities including secondary wastes from the fabrication of mixed oxide fuel. Work is also planned in conjunction with industrial partners to refine the process and secondary waste estimates already prepared for aqueous recycling of used fuel.

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