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A Characteristics-Based Approach to Radioactive Waste Classification in Advanced Nuclear Fuel Cycles

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A Characteristics-Based Approach to Radioactive Waste Classification in Advanced Nuclear Fuel Cycles

by

Denia Djokić

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Committee in charge:
Professor Per F. Peterson, Chair
Professor Brian D. Wirth
Dr. Kathryn A. McCarthy
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Abstract

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University of California, Berkeley
Professor Per F. Peterson, Chair

The radioactive waste classification system currently used in the United States primarily relies on a source-based framework. This has lead to numerous issues, such as wastes that are not categorized by their intrinsic risk, or wastes that do not fall under a category within the framework and therefore are without a legal imperative for responsible management. Furthermore, in the possible case that advanced fuel cycles were to be deployed in the United States, the shortcomings of the source-based classification system would be exacerbated: advanced fuel cycles implement processes such as the separation of used nuclear fuel, which introduce new waste streams of varying characteristics. To be able to manage and dispose of these potential new wastes properly, development of a classification system that would assign appropriate level of management to each type of waste based on its physical properties is imperative. This dissertation explores how characteristics from wastes generated from potential future nuclear fuel cycles could be coupled with a characteristics-based classification framework. A static mass flow model developed under the Department of Energy's Fuel Cycle Research & Development program, called the Fuel-cycle Integration and Tradeoffs (FIT) model, was used to calculate the composition of waste streams resulting from different nuclear fuel cycle choices: two modified open fuel cycle cases (recycle in MOX reactor) and two different continuous-recycle fast reactor recycle cases (oxide and metal fuel fast reactors). This analysis focuses on the impact of waste heat load on waste classification practices, although future work could involve coupling waste heat load with metrics of radiotoxicity and longevity. The value of separation of heat-generating fission products and actinides in different fuel cycles and how it could inform long- and short-term disposal management is discussed. It is shown that the benefits of reducing the short-term fission-product heat load of waste destined for geologic disposal are neglected under the current source-based radioactive waste classification system, and that it is useful to classify waste streams based on how favorable the impact of interim storage is on increasing repository capacity. The need for a more diverse set of waste classes is discussed, and it is shown that the characteristics-based IAEA classification guidelines could accommodate wastes created from advanced fuel cycles more comprehensively than the U.S. classification framework.

This dissertation is dedicated to my mother, Chris Schwarz, whose incredible sacrifices made everything possible.

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Acronyms

AIROX	Atomics International Reduction Oxidation
AEA	Atomic Energy Act
AEC	Atomic Energy Commission
Am	americium
C	carbon
Cd	cadmium
CFR	Code of Federal Regulations
Ci	curie
Cl	chlorine
COEX	Co-extraction (of uranium and plutonium)
CR	(transuranic) conversion ratio
Cs	cesium
DOE	Department of Energy
DU	depleted uranium
EA	Environmental Assessment
Echem	electrochemical
EIS	Environmental Impact Statement
EPA	Environmental Protection Agency
EU	enriched uranium
EW	Exempt Waste
FCT	Fuel Cycle Technology program
FIT	Fuel-cycle Integration and Tradeoffs
FP	fission product
FR	fast reactor
GSG	General Safety Guide
GTCC	Greater-Than-Class-C
GW	gigawatt
HEPA	High Efficiency Particular Air (a type of high-efficiency air filter)
HLW	high-level waste
HM	heavy metal (thorium, protactinium, uranium, or transuranics)
hr	hour
I	iodine
IAEA	International Atomic Energy Agency
iHM	initial heavy metal, the heavy metal content prior to irradiation of fresh fuel
ILW	intermediate-level waste
INL	Idaho National Laboratory
kg	kilogram
Kr	krypton
LLW	low-level waste
LWR	light-water reactor
MLLW	mixed low-level waste

MOX	mixed oxide fuel
MrTau	Multi-Reactor Transmutation Analysis
MW	megawatt
MWe	megawatt electric
MWth	megawatt thermal
NA	not applicable
NARM	Naturally Occurring Radioactive Materials or Accelerator-Produced Radioactive Materials
NCRP	National Council on Radiation Protection & Measurements
NORM	Naturally Occurring Radioactive Materials
Np	neptunium
NRC	Nuclear Regulatory Commission
NUEX	Neptunium (and Plutonium) extraction
NWPA	Nuclear Waste Policy Act
O&M	operation & maintenance
Pu	plutonium
PUREX	Plutonium Uranium Reduction Extraction
R&D	research and development
Ra	radium
RU	recovered uranium
Se	selenium
Si	silicon
SNF	spent nuclear fuel
Sr	strontium
Sv	Sievert
TALSPEAK	Trivalent Actinide Lanthanide Separation by Phosphoric Extractants and Aqueous Komplexes
Tc	technetium
TENORM	technologically enhanced NORM
Th	thorium
TRU	transuranic element (neptunium, plutonium, americium, curium, berkelium, californium)
TRUEX	Transuranic Extraction
U	uranium
UDS	undissolved solids
UOX	uranium oxide
UOX-51	uranium oxide fuel taken to a burnup of 51 MWth-day/kg-iHM
UREX	Uranium Extraction
UREX+	Uranium Extraction-plus
U.S.	United States
USC	United States Code
U-TRU	uranium-transuranic
VLLW	very low-level waste
VSLW	very short lived waste

WAC	waste acceptance criteria
WIPP	Waste Isolation Pilot Plant
Xe	xenon
YMR	Yucca Mountain Repository
yr	year
Zr	zirconium

Chapter 1

Introduction:
The Need for a Characteristics-Based Classification System for Radioactive Waste

Most processes in the nuclear fuel cycle—the activities associated with the production, utilization, and disposition of fuel for nuclear reactors—create wastes that are radioactive. These radioactive wastes differ in their compositions and concentrations of radionuclides, as well as in their chemical compositions and mobility. Different types of wastes have various potential impacts on human health and the environment. To minimize the impact on human health, radioactive wastes are managed and disposed of in a variety of ways. Depending on the waste, different levels of isolation from the biosphere are necessary, in time, space, or both. To efficiently determine how to protect human health from the hazards of radioactive waste, methods to classify these wastes must consider their hazards and how to reduce them. Various methods to classify these wastes have been developed in the United States and internationally over the last several decades. However, whether the resulting classification systems have all resulted in the most efficient and risk-minimizing outcome is not necessarily clear. This dissertation investigates the effectiveness of the radioactive waste classification system now used in the United States, and suggests potential improvements.

Generally, a radioactive waste classification system establishes a set of categories to group different wastes, each corresponding to waste management practices associated therewith. The criteria for classifying radioactive wastes are usually related to properties of the waste, such as its source, characteristics, potential risk to humans, or a combination thereof. [Croff 2006] Ideally, a radioactive waste classification framework should facilitate waste management and disposal practices by categorizing the wastes so they can be managed and safely disposed using appropriate technologies. Waste classification systems do not provide a substitute for site-specific assessments and waste-acceptance criteria (WAC) for waste management and disposal at individual facilities. Waste disposal facilities such as the Waste Isolation Pilot Plant (WIPP), the former Yucca Mountain repository (YMR) site, and various low-level waste (LLW) sites in the United States are required to have their own suitability criteria based on the physical properties of each individual site. [Croff 2006]

The radioactive waste classification system currently used in the United States primarily relies on a source-based framework. In other words, U.S. wastes are categorized by the processes in the fuel cycle from which they originate, and not by their physical properties or the specific hazards posed by their disposal. This approach is known to have many deficiencies [Lowenthal 1998]. For one, the basis of U.S. classification within the framework is inconsistent; for example, high-level waste is defined by its source, whereas low-level waste is defined by exclusion [10 CFR 61.55]. "By exclusion" means that low-level waste is defined as waste that is *not* a different class of waste, as further described in Section 1.2.3. Also, the classification rules leave some wastes "orphaned" without any legal framework for disposal. This includes mixed low-level waste (MLLW) and depleted uranium (DU) from enrichment plants, among others, which the regulatory framework does not currently address. Furthermore, the possible option of closing the fuel cycle, either partially or fully[1], involves processing used nuclear fuel to recover selected components for recycle, and therefore would lead to the creation of new waste streams from those processes. Under the current source-based framework, these would all be defined as high-level waste (HLW), although their characteristics and hazard levels may not reflect the need for HLW disposal requirements, or their physical properties may simplify and improve the performance of the disposal system.

The primary issues in using a source-based classification system is the inability to minimize both risk to the public and the disposal cost by assuring that all types of waste are managed optimally. Wastes from some sources may even be "orphaned," and excluded from disposal with other, similar materials. Suboptimal and even dangerous outcomes are possible, particularly due to the risk that orphaned wastes will be mismanaged and end up causing significant environmental, security, or public health problems. If one purpose of developing advanced fuel cycles is to reduce the long-term burden of radioactive waste, a source-based waste classification system also overlooks the potential benefits of fuel cycle options that would direct materials to be appropriately managed where they would generate the lowest risk to the public.

This chapter describes the current state of the radioactive waste classification system in the United States, contrasts this U.S. classification system with international approaches, and explores how the current U.S. classification system may not necessarily meet the purposes described above. It provides the background necessary to understand the need for a re-evaluation of the system, which is an inconsistent patchwork of source-based and characteristics-based criteria. Alternative attempts to reframe the radioactive waste classification system in the U.S., motivated by examples of classification frameworks used outside the U.S., are also discussed.

[1] Currently, the U.S. operates on a "once-through" nuclear fuel cycle, where the fuel is used only once and then treated as waste. Other countries, such as France, Japan, and the United Kingdom, implement a partially closed (also known as a "modified open") fuel cycle, where the uranium and plutonium in spent fuel is reprocessed once for recycle in reactors. "Fully" closing the fuel cycle would involve multi-recycle reactor systems where ideally, all actinides (or most, due to losses) would be recycled and only separated fission products from spent fuel would go to waste. [OECD 2011]

1.1. Legal and Regulatory Framework of the Radioactive Waste Classification System in the United States

Historically, the radioactive waste classification system in the United States has developed from a series of legal definitions for various types of radioactive wastes, which have generally been source-based. The focus has generally been on what processes generate the waste rather than the physical properties of the resulting waste or the subsequent effects on the environment and public health resulting from the ultimate disposition of the waste.

In the United States, the topmost statutory authority on which the radioactive waste classification is based is the Atomic Energy Act of 1954 (as amended) [AEA 1954]. The AEA, as amended, is the statute by which Congress delegates authority to federal agencies to manage and regulate nuclear materials and the disposal of nuclear waste. Initially, this federal agency was the Atomic Energy Commission (AEC), established in the first Atomic Energy Act of 1946, and which today has been split into two federal bodies, the Nuclear Regulatory Commission (NRC) and the Department of Energy (DOE). The policies and regulations that these agencies create must be consistent with Congress' statutes. [Lowenthal 1997] Among other directives, the AEA mandates the protection of public health and safety from the hazards of commercial nuclear power. [AEA 1954]

Over the years, Congress has passed additional legislation that provides the legal framework today for U.S. policy to manage nuclear wastes. The pieces of legislation and their impact on radioactive waste management are summarized in Table 1.1.

Table 1.1: Summary of legislation history contributing to the environmental protection from radioactive and hazardous wastes.

Legislation and year	Brief summary
National Environmental Policy Act of 1969, as amended [NEPA 1969]	Requires federal agencies to prepare Environmental Assessments (EA) and Environmental Impact Statements (EIS).
Energy Reorganization Act of 1974, as amended [ERA 1974]	Splits the function of development and regulation of nuclear technologies into the Energy Research and Development Administration (now DOE) and the Nuclear Regulatory Commission, respectively.
Resource Conservation and Recovery Act of 1976 [RCRA 1976]	Governs the disposal of solid and hazardous wastes.
Uranium Mill Tailings Radiation Control Act of 1978, as amended [UMTRCA 1978]	Amends the AEA 1954 to give the Environmental Protection Agency (EPA) authority to set environmental standards for disposal of uranium mill waste.
Comprehensive Environmental Response, Compensation, and Liability Act of 1980 [CERCLA 1980]	(Also known as Superfund.) Provides EPA the funds and the authority to clean up abandoned hazardous waste sites.
Low-Level Radioactive Waste Policy Act of 1980 [LLRWPA 1980] and the Low-Level Radioactive Waste Policy Amendments Act of 1985 [LLRWPAA 1985]	Sets responsibilities for individual States to dispose of low-level radioactive waste.
Nuclear Waste Policy Act of 1982 [NWPA 1982] and the Nuclear Waste Policy Amendments Act of 1987 [NWPAA 1987]	Makes States responsible for providing for the disposal of commercial LLW generated within their borders. Encourages States to enter into compacts that would allow several States to dispose of waste at a centralized regional disposal facility.
Energy Policy Act of 1992 [EPACT 1992]	Addresses sweeping energy efficiency, management, and conservation goals. Directs the EPA to promulgate radiation standards for Yucca Mountain Repository (YMR).

In 1974, under the Energy Reorganization Act of 1974, the Atomic Energy Commission was reorganized into the Nuclear Regulatory Commission (NRC), responsible for the safety regulation of the nuclear industry, and the Energy Research and Development Administration (now the Department of Energy (DOE)), responsible for the development and production of nuclear weapons and promotion of nuclear power. The NRC is charged with regulating commercial radioactive waste management activities but does not regulate defense HLW.

Significant quantities of HLW have been produced by the Department of Energy's defense reprocessing programs at facilities such as Hanford, WA and Savannah River, SC as well as by commercial reprocessing operations at West Valley, NY. Although the NRC does not regulate the production and storage of defense high-level wastes, it does regulate the licensing of civilian waste repositories that would co-mingle defense HLW [NWPA 1982].

1.2 Waste Classification Hierarchy and Characteristics

To a large extent, radioactive waste has been classified based on the potential radiation dose (current or future) that individuals could receive if exposed to the material. Comparison to background radiation levels has also been important in establishing negligible or acceptable levels of risk or dose, which in turn has, to a certain extent, informed the definition of waste classes. [Croff 2002]

The first level of distinction in the radioactive waste classification system occurs between wastes that are generated from nuclear fuel cycle activities versus other sources of radioactive waste (naturally occurring and accelerator-produced radioactive material, or NARM).

Fuel-cycle generated radioactive waste is produced in most processes of the nuclear fuel cycle: from mining, milling, fuel fabrication, reprocessing, to final disposal. For example, spent fuel is waste generated directly from irradiation in a reactor, and operational waste can a result of contamination of structural materials or personal protective equipment during a process in the fuel cycle. There are also wastes resulting from medical processes that are usually low-level. Fuel-cycle waste is currently broken up into spent nuclear fuel (SNF), high-level waste (HLW), transuranic waste (TRU), and low-level waste (LLW). Uranium mill tailings are classified as a byproduct material [10 CFR 40], but are nevertheless part of the nuclear fuel cycle.

Non-fuel cycle waste, or Naturally Occurring Radioactive Materials or Accelerator-Produced Radioactive Materials (abbreviated as NARM), can be further classified into subgroups of accelerator-generated waste and naturally occurring radioactive materials (NORM). The latter is divided up into regulated and unregulated NORM. "Technologically enhanced NORM" is known as TENORM and is a broader category that includes radioactive waste from technological and industrial processes. Examples of this include uranium mining waste, radioactive coal fly ash, and radioactive oil and gas drilling waste. Further than this, NARM waste is not discussed in this dissertation.

The current hierarchy of classes of radioactive wastes in the United States is illustrated in Figure 1.1, as illustrated in the 2002 NCRP report #139: Risk-Based Classification of Radioactive and Hazardous Chemical Wastes.

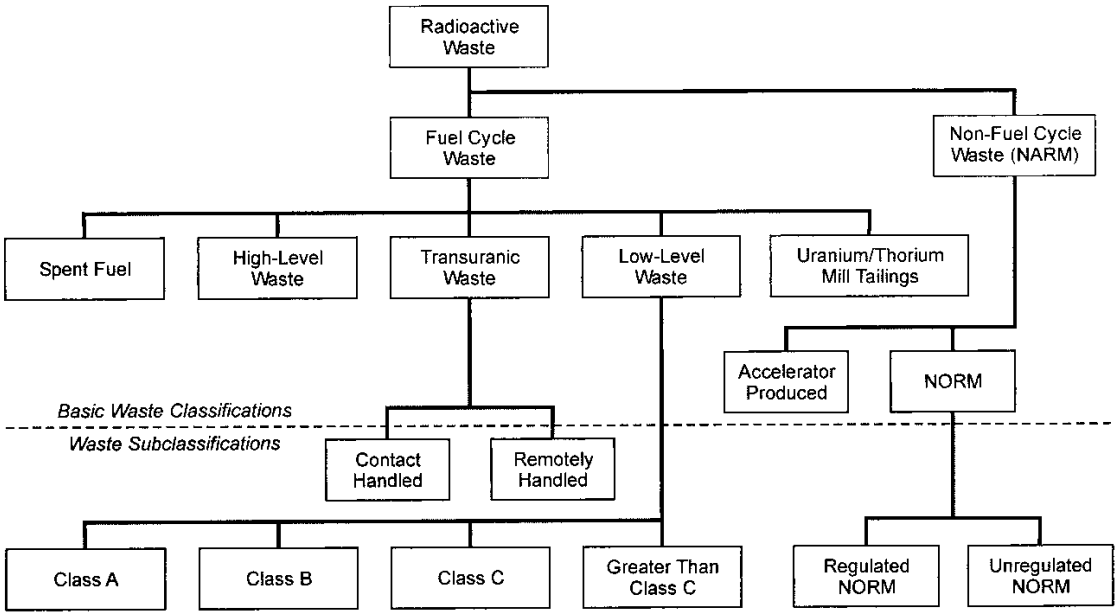

Figure 1.1: Current U.S. radioactive waste classification system [Croff 2002].

A general class for exempt wastes has not been established in the United States legal or regulatory framework. Currently, the fate of exempt wastes is determined on a case-by-case basis; in most specific instances, the NRC can exempt from regulatory control wastes that contain radionuclides in "sufficiently low concentrations" [42 USC 2021j]. No quantitative limit for exemption is given.

1.2.1 Spent Fuel and High-Level Waste

Under 10 CFR 60, spent nuclear fuel (SNF) is a subcategory of, and regulated as, high-level waste (HLW).

In the United States, the legal definition of spent fuel is "fuel that has been withdrawn from a nuclear reactor following irradiation, the constituent elements of which have not been separated by reprocessing" [10 CFR 60.2].

In the NWPA of 1982, high-level waste is defined as "(a) the highly radioactive material resulting from the reprocessing of spent nuclear fuel, including liquid waste produced directly in reprocessing and any solid material derived from such liquid waste that contains fission products in sufficient concentrations; and (b) other highly radioactive material that the Commission, consistent with existing law, determines by rule requires permanent isolation." [NWPA 1982]

In 1981, the NRC determined that irradiated reactor fuel shall, for the purposes of the repository, be considered HLW. Thus, 10 CFR 60—Disposal of High-Level Radioactive Wastes in Geologic Repositories explicitly includes spent fuel as high-level waste: "High-level radioactive waste or HLW means: (1) Irradiated reactor fuel, (2) liquid wastes resulting from the operation of the first cycle solvent extraction system, or equivalent, and the concentrated wastes from subsequent extraction cycles, or equivalent, in a facility for reprocessing irradiated reactor fuel, and (3) solids into which such liquid wastes have been converted." [10 CFR 60.2]

The entire classification system for fuel cycle wastes in the United States rests on this definition of high-level waste, because the definitions of transuranic and low-level waste depend on this definition. The general properties of high-level waste include high concentrations of shorter-lived fission products, resulting in high levels of decay heat and radiation (principally from Cs-137 and Sr-90, which have 30-year half lives), and a significant concentration of long-lived, alpha-emitting transuranic radionuclides, some of which generate significant heat over longer time frames (principally from Am-241, which has a 432-year half life, and Pu-238, with an 88-year half life). At present time, the NRC has chosen not to classify as high-level waste any waste that resembles high-level waste in its radiological properties but does not arise directly from the reprocessing of spent fuel.

Not only is the U.S. definition of high-level waste source-based, but it is also qualitative. Minimum concentrations, decay heat levels, and external radiation of fission products and long-lived, alpha-emitting transuranic radionuclides are not specified. This ambiguous definition has led to decisions by regulatory authorities to exclude some wastes that are part of fuel reprocessing but not directly resulting from the spent fuel itself, such as fuel cladding and ion-exchange beds, or salts produced in decontamination of liquid wastes. This ambiguity is further propagated in the definitions of transuranic waste and low-level waste. [Croff 2002]

1.2.2 Transuranic Waste

Transuranic, or "TRU" waste is a class of wastes that do not fall into the category of HLW, but contain "material contaminated with elements that have an atomic number greater than 92, including neptunium, plutonium, americium, and curium, and that are in concentrations greater than 10 nCi per gram, or in such other concentrations as the Nuclear Regulatory Commission may prescribe to protect the public health and safety." [42 USC 2014] This definition was revised in 1984 by DOE Order 5820.2 to be "without regard to source or form, waste that is contaminated with alpha-emitting transuranic radionuclides with half-lives greater than 20 years and concentrations greater than 100 nCi/g at the time of assay." [DOE 1988] TRU wastes mainly consist of clothing, tools, rags, debris, soil, and other items contaminated with transuranic elements. Most of the TRU inventory in the United States is legacy waste from decades-long nuclear weapons production and other defense-related research, and is managed by the Department of Energy.

The U.S. further categorizes TRU waste into contact-handled (CH) and remote-handled TRU waste, based on the radiation dose on the surface of the waste package, in order to inform packaging and handling requirements. If the measured dose rate is less than 2 mSv/h, it is categorized as contact-handled. If the dose rate is higher than 2 mSv/h and less than 10 Sv/h, it is categorized as remote-handled TRU waste. Defense TRU waste is managed by the DOE and can be placed into disposal in the Waste Isolation Pilot Plant (WIPP) facility, while no similar disposal method exists for civilian TRU waste (which mainly falls into the LLW GTCC category, as described below).

1.2.3 Low-Level Waste

As with defense TRU waste, in the U.S. low-level waste (LLW) is defined by exclusion. According to the NWPA of 1982, low-level radioactive waste is defined as "material that—

(A) is not high-level radioactive waste, spent nuclear fuel, transuranic waste, or by-product material as defined in section 11e(2) of the Atomic Energy Act of 1954 (42 USC 2014(e)(2));

and

(B) the Commission, consistent with existing law, classifies as low level radioactive waste." [NWPA 1982]

The "byproduct material" mentioned here refer to essentially uranium or thorium mill tailings. The LLRWPA defines low-level waste similarly, with the exception that it does not mention transuranic waste.

LLW is further subdivided into Classes A, B, C, and "Greater Than Class C" (GTCC). The categorization rules are set by the NRC in 10 CFR 61.55, Tables I and II, which apply characteristics-based rules based on the isotopic content of the low-level waste. [10 CFR 61.55] It is argued that these two tables are incomplete and do not consider some hazardous radioisotopes that could potentially be problematic in a disposal environment.

Most LLW has sufficiently low levels of radioactivity that most can be safely be placed in shallow land disposal. However, the "by exclusion" definition results in some waste materials that contain sufficient radioactivity to require geologic disposal, but cannot be categorized as HLW, to be assigned the class of GTCC low-level waste.

In the U.S., LLW generated by the DOE is managed differently from LLW generated by commercial and industrial sources. The DOE self-regulates its disposal of LLW, while for the commercial sector LLW disposal is regulated by the NRC under 10 CFR 61, based upon standards established by the EPA.

1.2.4 Uranium and Thorium Mill Tailings

Mill tailing wastes are residual wastes generated during the milling and chemical processing of ores of uranium and thorium. After mining and extraction, mill tailings are produced in large volumes and contain residual amounts of uranium or thorium that were not extracted during the milling process, as well as daughter products from the decay of uranium and thorium, which occur in relatively low concentrations but have very long half-lives. Mill tailings consist of "fine-grained, sand-like, and silty materials, usually deposited in large piles next to the mill that processed the ore" [OECD 2004]. In the United States, most tailing waste is disposed of near uranium mills, primarily located in western states such as Utah and Arizona. While the U.S. has some thorium deposits as well, thorium has never been actively mined in the United States.

The NRC requires licensees to meet EPA standards for cleanup of uranium and thorium mill sites after the milling operations have permanently closed. Mostly, mill tailings are disposed of by being capped and left in place.

Mill tailings share characteristics with other low-level, high-volume waste (e.g. depleted uranium and other generated waste from potential future fuel cycles, such as high-volume waste incidental to reprocessing). These wastes generate relatively low amounts of radioactivity but could contain significant levels of chemically hazardous heavy metals. It is important to note that in the U.S. mill tailings are regulated much differently from other types of low-level waste.

1.3 Deficiencies in the Current Classification System

The current U.S. classification system for nuclear wastes has several important deficiencies:

- Its classification criteria are inconsistent. For example, HLW is defined by its source, whereas TRU waste is defined by exclusion (see 1.2.2) and categorized into subclasses ("remote-handled" and "contact-handled"). LLW is also defined by exclusion (see 1.2.3) but then categorized into sub-classes depending on its characteristics, however, only for select isotopes (10CFR61 Tables I & II [10 CFR 61.55])[2].

- The definitions of some classes vary with the legal and/or regulatory framework; for example, the NWPA and the LLWRPA define LLW slightly differently (see 1.2.3).

- In some instances, similar waste is classified into separate categories. For example, some GTCC wastes are very similar to TRU wastes; however, since the former comes from a commercial source and the latter is waste originating from defense-related activities, they must legally be classified differently.

- There is no legal framework to deal with so-called "orphan wastes" for which there is no legal definition. These include mixed low-level wastes (MLLW), for which there is no facility to accept this waste outside the defense industry, irradiated material such as control rod blades, and sludges such as those resulting from cleanup of processing facilities (e.g. Hanford).

- Depleted uranium (DU) classification is controversial [Markey 2009]. In 2009, the NRC Commission voted to classify DU as Class A LLW; however, the large stockpiles and longevity of the depleted uranium (similar to mill tailings) make it difficult to deal with. Quantities of DU are now much larger than were considered in the Environmental Impact Statement (EIS) performed when 10CFR61 was written[3], and will be larger still with new enrichment facilities being licensed in the U.S. [Weinberger], enough to warrant disposal of (at least a portion of) the country's depleted uranium.

- There is no place for exempt wastes, i.e. wastes that are either not or negligibly radioactive and clear regulatory control after a very short period of decay time.

- The potential risk posed by radionuclides to human health is primarily mitigated by disposal design as opposed to upstream waste treatment choices.

Efforts to suggest a new classification framework, in the past (see Section 1.4) as well as in this dissertation, aims to address and eliminate these deficiencies.

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[2] Tables 1 and 2 of 10CFR61 are currently being revised to incorporate updated data and include problematic isotopes that have previously been ignored. [NRC 2008]

[3] 10CFR61 is currently being updated to better deal with DU [NRC 2008]. It recommends site-specific evaluation for DU disposal, depending on the quantities.

1.4 Efforts to Reclassify Radioactive Waste

Rethinking our radioactive waste classification system has been explored in the past [3]. Current revisions to 10CFR61 for low-level waste (LLW) definitions are being discussed, and on the international level, the IAEA recommends a characteristics-based classification system to its member states [4]. This dissertation aims to apply previous investigations of how to redraw the classification boundaries for radioactive waste to the results of the advanced fuel cycle systems model considered in this study.

1.4.1 IAEA Framework

The objective of the IAEA GSG-1 (General Safety Guide) [IAEA 2009] is to propose a general scheme for classifying radioactive waste that is based primarily on considerations of long term safety, and thus, by implication, disposal of the waste. The proposed classification system is based on IAEA safety standards that "reflect an international consensus on what constitutes a high level of safety for protecting people and the environment from harmful effects of ionizing radiation" [IAEA 2011].

This recommended classification system by the IAEA differs from that of the U.S. in several fundamental ways, including the following characteristics:

- It allows for a category of "exempt waste", which is defined as radioactive waste whose dose is negligible. This generally includes very short-lived isotopes, often from the medical field.

- It does not distinguish between wastes resulting from the nuclear fuel cycle and NARM. All materials are classified under the same system.

- It categorizes all wastes according to their properties rather than their source.

- Waste containing long-lived, naturally occurring radionuclides, such as uranium and thorium mill tailings, does not have its own waste class, whereas in the United States, it is recognized as a fuel cycle waste. Both classification frameworks identify that management and disposal of these high-volume wastes require special considerations.

The IAEA GSG-1 suggests six different classes of radioactive waste as the basis for a characteristics-based classification framework:

(1) Exempt waste[4] (EW): This category describes waste that "meets the criteria for clearance, exemption or exclusion from regulatory control for radiation protection purposes" [IAEA 2009].

(2) Very short lived waste (VSLW): This category includes waste that can be cleared from regulatory control by placing it into decay storage for up to a few years, after which it can meet the criteria for uncontrolled disposal. This class includes short-lived waste often used for research and medical purposes.

(3) Very low-level waste (VLLW): This class includes waste suitable for near-surface disposal (e.g. landfill-type facilities) with limited regulatory control. The radionuclides contained in this waste are generally short-lived, although very limited concentrations of longer-lived radionuclides are possible.

(4) Low level waste (LLW): This class covers a broad range of radioactive waste. It may include short-lived radionuclides at higher activity levels as well as long-lived radionuclides at relatively low levels of activity concentration. This waste requires containment and isolation for a longer period of time (a few hundred years) and usually is disposed of in engineered surface facilities.

(5) Intermediate level waste (ILW): This category of waste contain relatively large concentrations of long-lived radionuclides (generally alpha-emitting radionuclides) that will not decay to a low enough activity level during a reasonable period of time (i.e. a timeframe for which institutional control is not necessarily guaranteed). Waste in this class requires disposal at greater depths, tens or hundreds of meters below the surface.

(6) High level waste (HLW): Waste with levels of activity concentration high enough to generate significant quantities of heat by the radioactive decay process or waste with large amounts of long lived radionuclides that need to be considered in the design of a disposal facility for such waste. Disposal in deep, stable geological formations usually several hundred meters or more below the surface is the generally recognized option for disposal of HLW." [IAEA 2009]

Figure 1.2 presents a qualitative visual summary of these six suggested waste classes, categorized by their activity content (ordinate) and longevity (abscissa).

[4] The term 'exempt waste' is being used for consistency with previous suggested classification frameworks [IAEA 2004, Croff 2002]; however, it should be noted that it is not considered radioactive waste anymore after it has reached the threshold clearing it from regulatory control.

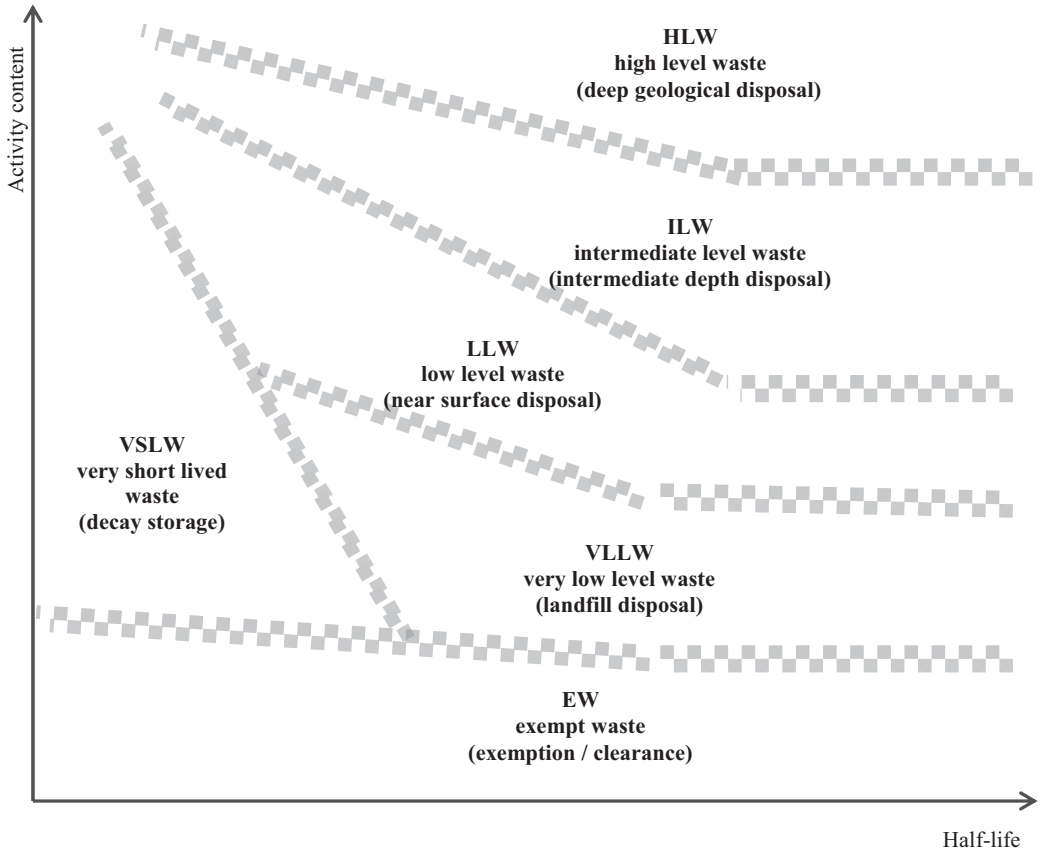

Figure 1.2: Conceptual illustration of the IAEA classification scheme [IAEA 2009]

The boundaries of the proposed waste classes are broadly shaded to indicate flexibility, and are not meant to indicate quantitative criteria. The guidelines state that it is the responsibility of each State (i.e. member country implementing a radioactive waste classification framework) to develop a set of quantitative criteria based on these recommended waste classes, which "may be presented in terms of levels of activity concentration, half-lives of the radionuclides in the waste, heat generated by the waste and/or dose or dose rate." [IAEA 2009] In addition to the characteristics of the waste, the disposal options available or under consideration in each State also influence the exact criteria used to assign waste to a particular class.

In the IAEA classification framework, waste heat generation is not explicitly used as a classification criterion; it correlates with half-life, decay energy, activity concentration, and total activity. Furthermore, management of decay heat (e.g. ventilation, thermal conductivity of host geology or storage matrix) is expected, if needed. Therefore, the IAEA framework states that heat generation cannot be defined by a single parameter.

1.4.2 Additional Classification System Options

A classification system first suggested by Kocher and Croff in 1987 [Kocher 1987], and then revised in 2002 [Croff 2002], suggests a risk-informed classification system based on the characteristics of the waste. The framework posits categories to cope with with radioactive *and* chemical wastes based on an intrinsic hazard to the public. The qualitative descriptions of the three proposed subclasses shown in Figure 1.3 (exempt waste, low-hazard waste, and high-hazard waste) link the waste categories to the risk presented by the capability for waste isolation of the disposal system.

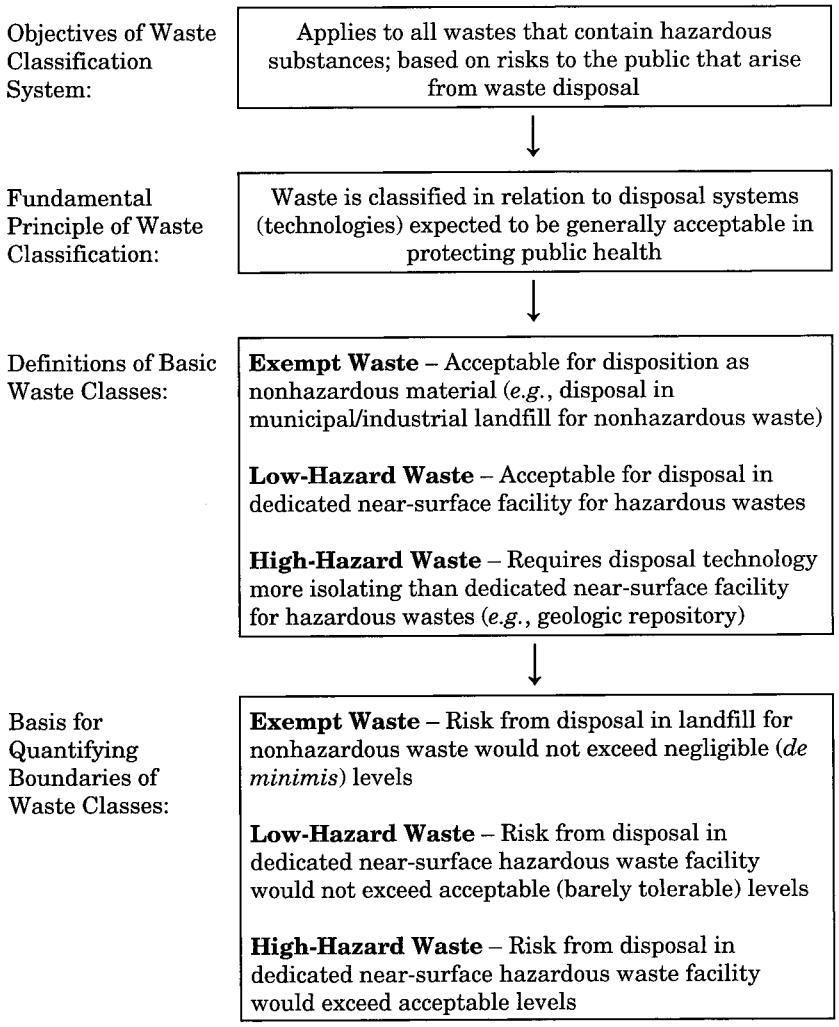

Figure 1.3: Basic framework for NCRP report #139 proposed classification system. [Croff 2002]

Similarly, Smith and Cohen [Smith 1989] propose a classification framework that associates disposal technology with the waste class. The four proposed classes of radioactive waste are BRC (below regulatory concern) waste, low-level waste, intermediate-level waste, and high-level waste. No detailed risk analysis is performed in this study, but some concentrations limits for waste class boundaries are suggested.

A proposed classification framework developed by LeMone and Jacobi [LeMone 1993] suggests the same four classes of radioactive waste as Smith and Cohen: BRC (below regulatory concern) waste, low-level waste, intermediate-level waste, and high-level waste. In contrast to other suggested classification systems, the first three suggested classes are found to be acceptable for near-surface geologic disposal, whose isolation would depend on different engineered barrier systems. It is proposed that waste class boundaries be quantified by imposing annual dose limits to maximally exposed individuals. No radionuclide-specific concentration limits are suggested.

Several studies have been done for re-evaluation of low-level waste classification that address the deficiencies of Tables I and II in 10 CFR 61 [NRC 2008]. For one, Lieberman and Greeves [Lieberman 2011] suggest a number of risk-informed, performance-based rules: requiring site specific performance assessment for both the public and intruder scenarios; setting a time of compliance for such calculations; setting a specific intruder dose standard; use of modern International Commission on Radiological Protection (ICRP) dose conversion recommendations; requiring periodic updates to the performance assessment to account for unique waste streams and improved site specific knowledge; upgrade compatibility requirements for 10 CFR 61.58; and stating in 10 CFR 61.55 the point in time for classifying waste.

In general, all of these existing reclassification proposals are similar to the International Atomic Energy Agency's General Safety Guideline recommendations on classification. All proposed frameworks recognize the association of waste classes with available disposal systems; most include a class of exempt waste or equivalent, and to a certain extent suggest quantitative boundaries between different waste classes.

1.5 The Need for a New Classification System

The 1982 Nuclear Waste Policy Act established a fee charged to U.S. utilities to fully fund the federal government's effort to place spent fuel into disposal. The fee is assessed as a charge on nuclear electricity generation, and was set to be 0.1 cents per kilowatt-hour. [NWPA 1982] The Secretary of Energy is required to review the fee annually, and today the fee remains the same. Electricity generation is an indirect and imperfect metric for nuclear waste generation. For reactors with the same thermal efficiency, it is proportional to the total inventory of spent fuel, but it does not reflect significant changes that have occurred in fuel burnup, with resulting alteration of the mass of spent fuel and transuranic content over the last three decades. Likewise, for advanced, non-water cooled reactors such as high-temperature gas cooled reactors and sodium fast reactors, actual spent fuel composition will differ greatly when evaluated on the basis of net electricity generation, and the fee structure provides no credit for the effects of reducing waste production or reprocessing.

A reformed waste classification system could result not just in better nuclear waste management and disposal practices, but also provide better price signals to nuclear waste generators to encourage efforts to reduce waste generation. In particular, changes to the radioactive waste classification system would provide a better basis to implement advanced fuel cycles to reduce the long-term impacts of nuclear waste, so the waste classification system would consider the advantages provided by different advanced fuel cycle options (i.e. reducing the inventory of what is considered hazardous) and maximize the benefits of the fuel cycle by directing materials to a management strategy where they would cause the lowest risks.

As discussed earlier, several proposals for reclassifying radioactive wastes have been discussed in the past. All argue that a characteristics- and risk-based approach to classifying radioactive wastes make more sense, but none take into account the imperative of rewriting our classification system if the United States were to deploy advanced fuel cycles which involve the partitioning of spent fuel. Ideally, the classification system should be fuel-cycle neutral, but further work is needed to understand the details of what this would look like.

A possible expansion of nuclear energy to advanced fuel cycles would involve the processing of spent fuel and the creation of new and very different waste streams. The shortcomings of the current classification framework would be exacerbated in most future fuel cycle scenarios. Thus, this investigation aims to suggest a new framework for classifying radioactive waste that is independent of fuel cycle choice and based on waste stream characteristics.

In general, a radioactive waste classification system should meet the following criteria:

- **Optimize between simplicity and diversity**: A classification system should be simple enough to be cost effective, but diverse enough to meet risk requirements.

- **Be "system-neutral"**: It should apply to any set of wastes from any choice of fuel cycle. The IAEA aims to make its classification guidelines universally applicable.

- **Be internally consistent**: Definitions of different wastes should be based on the same metric for fair comparison; ideally, criteria should be characteristics-based and incorporate impact on human health and environment. The current classification system is made up of varying waste class definitions with inconsistent bases for comparison.

- **Inform R&D prioritization**: An integrated waste management strategy requires knowledge of how upstream and downstream processes in the fuel cycle connect. The current classification system presents an overly rigid framework that is not sensitive to upstream changes in the fuel cycle, and there is no way to evaluate the potential benefits thereof and where R&D efforts need to be focused.

- **Adapt to societal risk (short- and long-term)**: A waste classification system should be able to intrinsically minimize risk to the public. This includes recognizing how wastes are handled prior to disposal based on their characteristics (e.g. classify wastes into "contact-handled" and "remote-handled", as in TRU definition). The current classification system pays lip service to the protecting the public from hazardous material, but has so many deficiencies that it fails to do so on many levels.

- **Ensure no orphan wastes, and have a place for exempt wastes**: In the past, policy has been written based on the type, source, and amount of waste at the time. Possible future developments (in technology, policy, processes) were not considered, leading to orphan waste that fall under no specific definition. This can be avoided by simply being allowed to categorize a waste by what it looks like and how hazardous it is, not by where it came from. The current classification system has no comprehensive way of dealing with orphan wastes.

- **Encourage good waste management practices prior to disposal**: It is usually necessary to have appropriate treatment and disposal strategies for each specific type of waste to minimize risk to the public. The current classification system does specify a disposal technology corresponding to a certain type of waste in some instances, but does not consistently do so.

- **Lead to appropriate and cost effective disposition**: Overall, waste treatment and disposal cost is a modest fraction of the entire fuel cycle, but ensuring that wastes are treated and disposed of properly ensures efficient allocation of financial resources.

The study presented in this dissertation briefly touches on some of these points but explores others in more depth, as summarized below.

1.6 Scope of the Waste Reclassification Study

The goal of the study presented in this dissertation is to provide recommendations on how advanced fuel cycle wastes can inform and strengthen the case for a characteristics-based classification framework. Although a classification system should be independent of the choice of fuel cycle and should ideally meet all criteria listed above, the scope of this study does not encompass addressing all of the points presented above. Although briefly touched upon, this study does not focus on exploring the cost and economics of waste disposal, informing R&D prioritization, or directly minimizing risk to the public. This study addresses the problem of orphan wastes and informing waste management practices due to waste characteristics, based on technical evaluation of advanced fuel cycle waste stream characteristics. The objective of this study is to explore how key waste characteristics change when different fuel cycle options are used, and to demonstrate how the current U.S. waste classification framework is not sufficient to handle these wastes. Also, this study aims to inform waste management practices due to heat content of different wastes. Furthermore, it is shown that a characteristics-based framework such as that suggested by the IAEA is more effective in addressing issues of orphan waste and classification criteria consistency.

Chapter 2 presents a fuel cycle modeling tool, FIT (Fuel-cycle Integration and Tradeoffs), that tracks and compares mass flows from advanced fuel cycle systems and evaluates the composition of wastes from various fuel cycles, based on most current available data. It allows key waste characteristics, such as decay heat generation, radiotoxicity, mass, and volume to be quantified.

Chapter 3 discusses the results of FIT-generated cases and analyzes how advanced fuel cycle waste streams would be classified under the current classification system and what the deficiencies of doing such are. Chapter 3 examines what waste characteristics can be used as metrics to make prudent waste management decisions, and how a characteristics-based approach to a classification system can maximize the benefit of advanced fuel cycles. To illustrate the application of fuel cycle modeling to predict waste characteristics, a detailed study of waste decay heat generation characteristics is presented. Chapter 3 then discusses other metrics for waste classification, such as radiotoxicity. Suggestions on how this would inform a characteristics-based classification system are presented. The case is made for a need for a characteristics-based classification system to efficiently manage radioactive waste from any part of any fuel cycle of choice.

Chapter 4 summarizes the dissertation, highlights the main discussion points, and discusses future work needed to develop a characteristics-based waste classification system.

Chapter 2

Tools and Methodology Framework: The Fuel-cycle Integration and Tradeoffs Model

In order to study the characteristics of wastes generated by advanced fuel cycles, a fuel cycle tool that provides a comprehensive choice of possible future nuclear fuel cycle scenarios and a detailed data set of the corresponding wastes generated was needed. The tool of choice in this study is the Fuel-cycle Integration and Tradeoffs (FIT) model, which provides the basis of the methodology framework for the nuclear fuel cycle analysis in this study. The purpose of the model and its structure are described in this chapter. The selection process of representative use cases is explained. The deficiencies of the model and future model development work are also clarified. Assumptions made are discussed where applicable.

2.1 FIT Model Overview: Description, Purpose, and Capabilities

The Fuel-cycle Integration and Tradeoffs (FIT) 2.0 model, developed by the Systems Analysis Campaign at Idaho National Laboratory (INL), is a static mass flow model that tracks isotopes through a fuel cycle of the user's choice; from discharging used fuel, to fuel recycling in a reactor, to waste disposition [Piet 2012]. FIT is a systems-level model that allows the user to compare nuclear fuel cycles with different parameters of choice, and to analyze how changes in these parameters in one part of the fuel cycle affect other parts of the fuel cycle. The nature and value ranges of these parameters are discussed in this chapter.

The two key capabilities that motivated the development of the FIT model were: (1) estimation of impurities fabricated fuel and losses of unwanted isotopes in waste as a function of fuel cycle parameters; (2) identification of whether a certain fuel cycle scenario is feasible based on fuel cycle technology choices, fuel cycle parameter choices, and the potential accompanying impurities. These initial motivations led to the development of a tool that can perform mass tracking through an entire advanced nuclear fuel cycle system.

Figure 2.1 depicts the basic mass flow and processes of the FIT model. The incoming light water reactor uranium oxide (LWR UOX) used fuel at an assumed, nominal burnup of 51 MWth-day/kg-iHM that is typical of current LWR operation, goes through an aqueous separation process. The recovered actinides, the recovered uranium, and depleted uranium (DU) as a makeup feed, provide the material needed to fabricate metal or oxide fuel for irradiation in a second reactor, either for multiple cycles in a fast burner reactor, or one cycle in a mixed-oxide fuel light water reactor (MOX-LWR), where the discharged MOX fuel goes to waste and no minor actinides are recovered. After discharge in the fast reactor cases, the used fuel goes through a second separation: electrochemical reprocessing if a metal fuel is used in the recycle fast reactor, and an aqueous process if an oxide fuel is used. Some materials are recovered for another pass in the reactor and the rest are managed as waste. FIT is capable of modeling up to 19 recycle loops through the recycle reactor, in appropriate cases. In the two fast burner cases in this study, 19 recycle iterations are sufficient for the fuel to approach equilibrium composition values, and do not affect fuel fabrication impurity limits. However, for MOX cases, only one recycle is possible due to accumulated impurities and thermal limits due to heat-generating actinides.

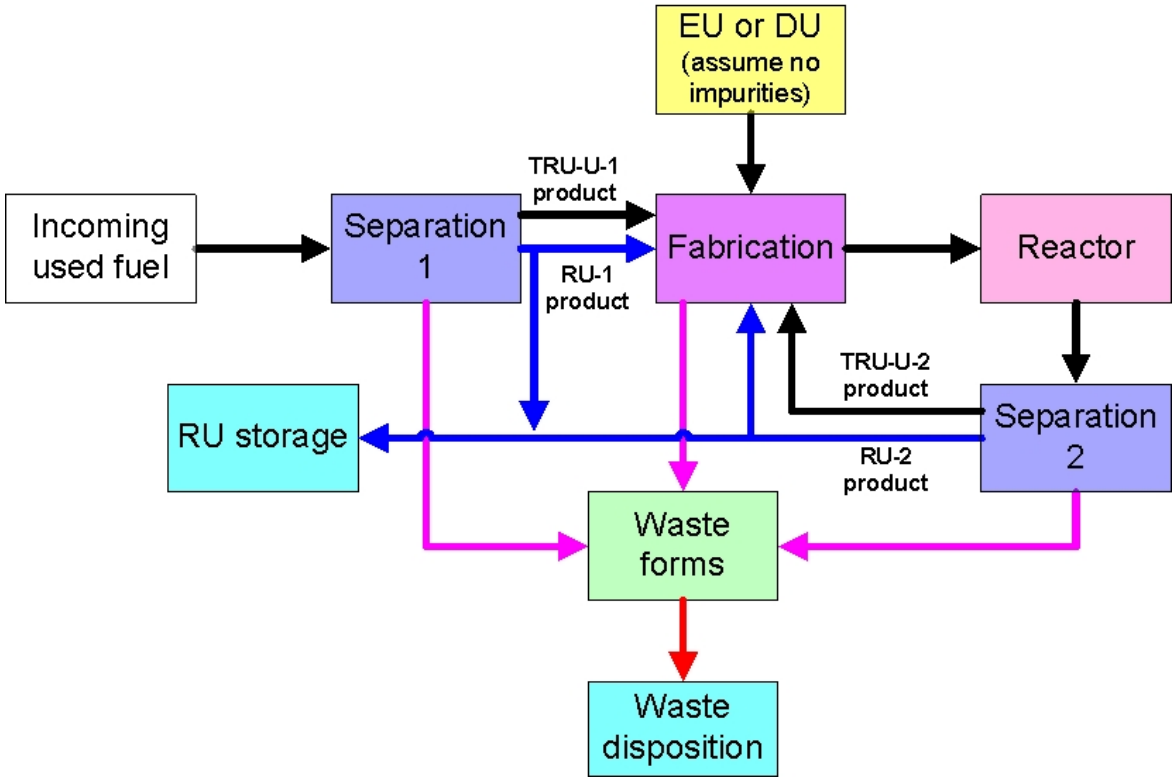

Figure 2.1: Basic mass flows; FIT 2.0 does not yet address EU feed as an option.

For every case in this study, recovered uranium RU-1 was used for the first iteration (RU-2 is not generated unless the first cycle is completed, as shown in Figure 2.1), and RU-2 was used for every recycle after the first. RU-2 and recovered transuranic elements (TRUs) are kept together for fuel fabrication. No breeder cases were considered for this study. Therefore, every case considered has excess uranium in the system; so it is reasonable to assume that there will always be enough RU-2 as a makeup feed for fuel fabrication.

There are two separations modules: Separation 1, in which the partitioning of the incoming used fuel (LWR UOX-51) occurs, and Separation 2, where the used fuel of the second reactor is processed. For each of these separation modules, FIT allows for several separation technology options, but for this study, only UREX+1a, electrochemical processing, and PUREX were considered where appropriate. Section 2.1.1.2 describes each of these separation processes and how each option manages the minor actinides (Np, Am, and Cm).

For simplicity, the separation efficiencies for each module are assumed to be constant with every recycle. The user can define a loss fraction for undissolved solids (UDS) in aqueous separations processes. For the base case of this study, a 0.1% UDS loss fraction was assumed for UREX+1a separations, although the sensitivity of output parameters as a function of this loss fraction is explored in Section 3.3. In cases of when UREX+1a is used (i.e. all cycles of oxide fuel fast reactor case and the first cycle of metal fuel fast reactor case), the undissolved solids and therefore its corresponding (non-ECHEM) metal alloy ingot waste form have the same fractional composition as the used fuel. The user can also choose whether UDS gets vitrified along with the raffinate/residual waste in HLW glass for aqueous separations processes, or stays as a separate waste stream to be immobilized in a metal alloy ingot. In this study, UDS was assumed to combine with the HLW glass waste form. For an electrochemical separations process, UDS, cladding, and baskets are immobilized in a metal alloy ingot.

Waste management is addressed in the model by tracking the masses of all materials separated from used fuel that are not recycled into new fuel. Waste streams generated during fuel fabrication are also tracked. All tracked waste streams are converted into a waste form suitable for disposal, based on current waste form feasibility studies [Gombert 2007]. The waste stream characteristics can then be examined to make assessments about their management. At this time, FIT does not calculate waste volumes based on waste packaging design assumptions. This was done to avoid confusion with the actual volume of the waste forms.

No operational and maintenance (O&M) waste from front-end processes, the initial fuel fabrication, or the two reactors in the cycle (the initial LWR and the recycle reactor) have been considered in the model implementation to date. However, O&M wastes for separations and fuel fabrication are included; FIT currently only deals with “technology wastes” that specifically contain wastes separated from the used fuel. See Section 2.2.4 for a more detailed discussion of O&M waste.

2.1.1 Description of FIT Modules, Parameter Options, and Assumptions

This section describes the function of each module in the FIT model, and lays out the parameter options any FIT user has. Assumptions made are also discussed.

Figure 2.2 shows the set of parameter choices the user has (in green) and the mass flow paths (red). FIT modules (black) are shown with their respective input and output variables. Note: This is from the version of FIT that does not have the flexibility to "smart"-classify the output waste, so it is drawing on existing classification definitions.

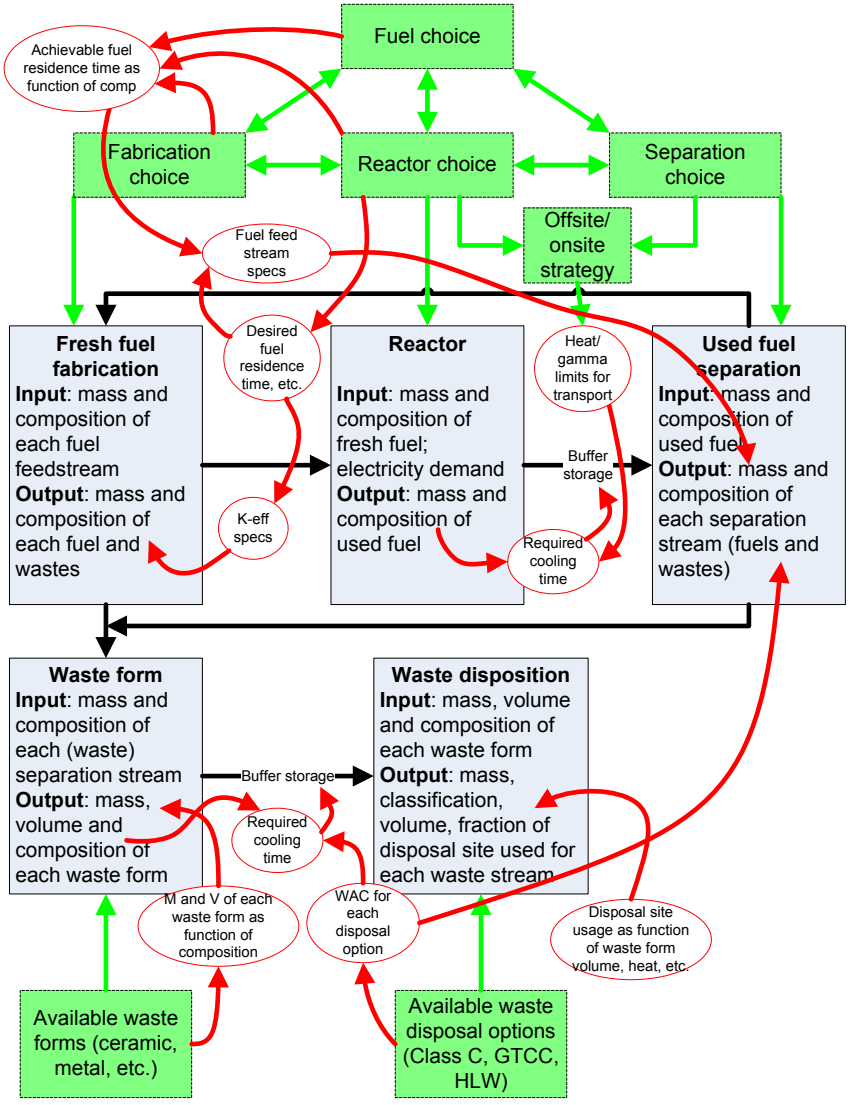

Figure 2.2: Technological choices (green) and information flows (red) overlaying mass flows (black). [Piet 2012]

2.1.1.1 Input Feeds

The default input feed for any fuel cycle model chosen in FIT is a feed of LWR UOX at 51 MWth-day/kg-iHM burnup. [Stillman 2004] There exists flexibility in FIT to replace the incoming raw data with another used fuel type, but for simplicity different input feed options were not considered in this analysis.

The recovered uranium (RU) stream from each of the separations modules, Separation 1 and Separation 2, illustrated in Figure 2.1, is labeled RU-1 and RU-2, respectively. In every case in this study, RU-1 was used for the first iteration (RU-2 is not generated unless the first cycle is completed), and RU-2 was used for every subsequent iteration (keeping RU-2 and TRU together for fuel fabrication). Note: of the cases considered in this study (see Table 4.1), none incorporate breeder reactors into the fuel cycle. Therefore, there will always be more RU-2 than is needed for the makeup fuel for the second reactor, so it is (reasonably) assumed that there will always be enough RU-2 as a makeup feed for fuel fabrication. If there is ever a case where there is not sufficient RU-2, FIT supplements the RU needed by RU-1.

In the current default input configuration for depleted uranium (DU) should this be chosen, the composition of DU values assumes a concentration 0.2% U-235 which is a typical enrichment tails assay and no impurities. Figure 2.1 shows that there is a DU or EU (enriched uranium) feed option for fuel fabrication; however, the option to add EU as an option is currently still in development.

2.1.1.2 Separations

There are two separations modules: Separation 1, in which the reprocessing of the incoming used fuel (LWR UOX-51) happens, and Separation 2, where the used fuel of the second reactor is separated. For each of these modules, there is an option of selecting among the following separation technologies: UREX+1a (FCT basis), PUREX, UREX+1a (PUREX basis), electrochemical (ECHEM), AIROX (minimum fuel treatment separation), melt refining, and fluoride volatility [Piet 2010b].

For simplicity, the separation efficiencies for each isotope for each separation module are assumed to be constant with every recycle. (In reality, as impurities accumulate with each recycle loop, separation efficiencies for different isotopes might decrease.) In cases where no separation data were available, 99.9% isotopic recovery fraction was assumed as a default. The user is also able to set the separation matrix manually in the FIT program. For UREX+1a processes, when applicable, the user is able to choose a loss fraction of the separation stream to the undissolved solids (UDS) stream.

Alternative separations processes such as AIROX, fluoride volatility, and melt refining were not considered in this study. The separations processes that are relevant for this study (PUREX, UREX+1a, ECHEM) are described in this section. While there exist other separations technologies such as NUEX, which is similar to UREX+1a with more flexibility, and COEX, which is a form of the PUREX process that co-extracts U and Pu [PATEROS 2008], the extraction processes chosen in this study are representative of three main types of technologies used in radioactive waste reprocessing.

In FIT, there is an option to perform a storage decay calculation after with each separations step (Separation 1 or 2) that can be utilized as desired. For this study, the idealized assumption was made that there is no storage time between processes, with the exception of the initial storage time before Separation 1, corresponding to the feedstock age. Storage time after Separation 1 (the number of years between separations 1 and when fuel is blended and inserted into the reactor), Reactor storage (the number of years between reactor discharge and when fuel is enters separation 2), and storage after Separation 2 (the number of years between separations 2 and when fuel is blended and inserted into the reactor), are all assumed to be zero.

2.1.1.2.1 PUREX: Plutonium-Uranium Reduction EXtraction

The PUREX method is an aqueous separation process used to recycle light water reactor fuel. Plutonium and uranium are extracted for re-fabrication into mixed-oxide (MOX) fuel for recycle in thermal reactors. PUREX is the standard liquid-liquid extraction method used in most countries that reprocesses used commercial nuclear fuel, such as France or the United Kingdom. The PUREX process leaves the minor actinides (Np, Am, and Cm) in the fission product waste stream.

In the FIT cases considered in this study, PUREX is appropriate for application in the LWR fuel reprocessing MOX cases.

2.1.1.2.2 UREX+1a: URanium EXtraction

UREX+1a, also an aqueous separations process, is one of the options of the UREX+ suite, a collection of aqueous separation technologies, developed at Argonne National Laboratory. The UREX+ suite is shown in Table 2.1, and a more detailed visualization of the UREX+1a option is shown in Figure 2.3. UREX+1a is the option in the suite that separates a pure TRU stream, which includes the minor actinides, from the fission product stream. This enables recycle in fast reactors. The UREX+ data used for the FIT cases in this study are based on the data from the Fuel Cycle Technologies (FCT) investigations of the UREX+ process, which involves just one separations cycle (in contrast to PUREX's three cycles, as in France). [Piet 2011a]

Table 2.1: Suite of UREX+ processes. [Laidler 2007]

Process	Product #1	Product #2	Product #3	Product #4	Product #5	Product #6	Product #7
UREX+1	U	Tc	Cs/Sr	TRU+Ln	FP		
UREX+1a	U	Tc	Cs/Sr	TRU	All FP		
UREX+2	U	Tc	Cs/Sr	Pu+Np	Am+Cm+ Ln	FP	
UREX+3	U	Tc	Cs/Sr	Pu+Np	Am+Cm	All FP	
UREX+4	U	Tc	Cs/Sr	Pu+Np	Am	Cm	All FP

Notes: (1) In all cases, iodine is removed as an off-gas from the dissolution process.
(2) Processes are designed for generating no liquid high-level waste.

U: uranium (removed to reduce the mass and volume of other streams)
Tc: technetium (long-lived fission product, prime contributor to long-term dose at Yucca Mountain geologic repository)
Cs/Sr: cesium and strontium (primary short-term heat generators; repository impact)
TRU: transuranic elements (Pu: plutonium, Np: neptunium, Am: americium, Cm: curium)
Ln: lanthanide (rare earth fission products)
FP: fission products other than cesium, strontium, technetium, iodine, and the lanthanides

The assumptions made to incorporate UREX+1a functionality in the FIT model are as follows: Most of the separation factors for the UREX+1a scheme are based on [Pereira 2010]. No separation factors for Cf or Bk were given, so the separation factors for Cm were used. It was assumed that the actinides lost during UREX separations all combine with the uranium stream, although in reality, a fraction could have gone to the UDS or Tc stream.

For lanthanide separation in any UREX+1a case, the user has the option to combine Ln with HLW glass form. If this option is chosen, then lanthanides are lumped in with the raffinate/residual waste in the HLW glass form. If not chosen, the lanthanides would go into a separate lanthanide borosilicate glass waste form. Such a waste form has a higher waste loading, and therefore higher heat load. Separating lanthanides was once considered as a realistic option in UREX+ waste management strategy, but now it is generally accepted that lanthanides are just left in FP waste stream due to tradeoffs with cost and repository footprint [Soelberg 2012]. A parametric study (Section 3.3) explores the sensitivity of this choice on the final results of this study.

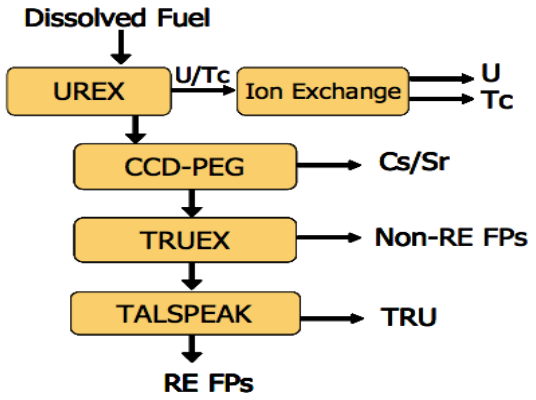

Figure 2.3: Schematic of the UREX+1a separations process and its constituent separation modules.

2.1.1.2.3 ECHEM: Electrochemical Separation

Electrochemical separation (ECHEM), commonly called pyro-reprocessing, is a dry chemical separation process that is well suited for recycle of fast reactor fuels using base salts and electrorefiners. The technique, while not yet widely used, is useful in treating fast reactor fuels due to its ability to handle higher burnups because there is no solvent radiation damage as opposed to aqueous processes. [Goff 2011] Electrochemical separations processes have been shown to have the ability to dissolve and separate up to 99.7% of actinides [Li 2005], which lends itself well to recycle into new fuel, and results in significant benefits of reducing the actinide burden on repositories.

Electrochemical processing is not as ideal for partitioning of fuel from thermal reactors because it does not remove enough of the fission products, which act as a neutron poison in the reactor. As a result, in FIT, even electrochemical cases still use UREX+1a to treat the incoming LWR-UOX fuel. Within the framework of this study there is always a need for an aqueous reprocessing scheme in the fuel cycle, regardless of choice of fast reactor fuel.

This technology has been used in the dry process for treatment of used fuel from the Experimental Breeder Reactor II (EBR-II) at Idaho National Laboratory. The electrochemical separation data incorporated in the FIT model are estimates based on experience treating EBR-II spent fuel, small-scale experiments, and expected chemical behavior.

2.1.1.3 Fuel Fabrication

After the first separation (Separation 1) of LWR-UOX fuel, recovered fuel is sent to the fuel fabrication module, where it is refabricated into either MOX fuel or metal or oxide fast reactor fuel. For cases with multiple recycles, fuel is refabricated as often as needed after the second separation (Separation 2). The FIT model calculates impurity levels of the refabricated fuel, and the user determines if these levels are acceptable and how input parameters must be changed to achieve the desired purity of the fresh fuel.

2.1.1.4 Storage

Decay calculations are built in throughout the model [Bays 2010, Piet 2011b]. The user has the option of choosing storage time (in years) to be calculated at the following stages of the FIT model: after initial used fuel discharge and before first separation, after first separation and before fuel fabrication, after fuel fabrication before insertion into the reactor (assumed to be zero in all calculations), before the second separation after a pass in the reactor, and finally, decay storage for appropriate waste forms (e.g. Cs/Sr or Kr/Xe).

2.1.1.5 Reactor

Currently, the user has several choices for reactor types and associated parameters in FIT. These are summarized in Table 2.2.

Table 2.2: Reactor cases pre-loaded in FIT. [Piet 2012] See Acronyms for key to acronyms. The fuel time residence in the recycle reactor does not vary significantly with multiple recycles.

Recycling reactor	Neutron Spectrum	Aging of feedstock from LWR UOX (yr)	Type of uranium used	Burnup (MWth-day/kg-iHM)	Average residence time of fuel (yr)
FR metal CR=0.50, startup, from 5yr-old LWR UOX	Fast	5	RU	131.9	4.44
FR metal CR=0.50, equilibrium, from 5yr-old LWR UOX		5			
FR oxide CR=0.50, startup, from 5yr-old LWR UOX		5		166.0	6.58
FR oxide CR=0.50, equilibrium, from 5yr-old LWR UOX		5			
FR metal CR=0.75, startup, from 5yr-old LWR UOX		5		99.6	4.68
FR metal CR=0.75, equilibrium, from 5yr-old LWR UOX		5			
FR oxide CR=0.75, startup, from 5yr-old LWR UOX		5		130.9	7.10
FR oxide CR=0.75, equilibrium, from 5yr-old LWR UOX		5			
MOX-Pu studies, from 5yr-old LWR UOX	Thermal	5		50	5.00
MOX-NpPu studies, from 5yr-old LWR UOX		5			
MOX-Pu studies, from 10yr-old LWR UOX		10	DU	51	4.56
MOX-NpPu studies, from 10yr-old LWR UOX		10			
MOX-NpPuAm studies, from 10yr-old LWR UOX		10			
MOX-TRU studies, from 10yr-old LWR UOX		10			
MOX-Pu feedstock aging, from 10yr-old LWR UOX		10	RU	50	5.00
MOX-NpPu feedstock aging, from 10yr-old LWR UOX		10			
MOX-Pu feedstock aging, from 20yr-old LWR UOX		20			
MOX-NpPu feedstock aging, from 20yr-old LWR UOX		20			

MOX-Pu feedstock aging, from 50yr-old LWR UOX		50			
MOX-NpPu feedstock aging, from 50yr-old LWR UOX		50			

In the above list of reactor choices, note that selecting a reactor case also determines whether the fuel of that reactor is metal or oxide fuel.

The FIT user can choose either "startup" or "equilibrium" system conditions for the recycle system. A "startup" reactor choice means that the reactor is in equilibrium with the material coming directly from the first recycle. In this study, "startup" reactor cases have been selected for the first recycle of all FIT cases run. An "equilibrium" reactor system means that the recycled material composition has already stabilized. In this study, "equilibrium" cases were selected for fuel cycle cases in which material is recycled many times (i.e., fast reactor cases after the first recycle). The main difference between the "startup" and "equilibrium" cases is the actinide mix, which will vary somewhat; mostly there is a larger concentration of high-Z TRU isotopes in equilibrium composition cases (for non-breeder cases). But, from the standpoint of waste management and impurity accumulation, it is the fission product yield that matters and that changes very little as the actinide mix varies. So, for the purposes of this study, there is generally little difference in FIT calculations if one uses "startup" versus "equilibrium" compositions.

The FIT model's reactor module calls a Fortran executable called MrTau, a depletion model similar to ORIGEN that calculates output fuel composition [Alfonsi 2011]. For this study, all U/TRU fast reactor choices are run with a transuranic conversion ratio (CR) ≤ 1. Simulations with CR > 1 (i.e. breeder reactors) was not considered. Thus, an incoming used fuel feed is always needed for makeup fuel in the system.

For the first recycle iteration, RU-1 was chosen as the RU makeup feed (there is no RU-2 for the first recycle) and RU-2 for subsequent recycles, corresponding to keeping all the U with TRU in fast reactor recycling. In the current version of FIT, the supply of the selected source of uranium is unlimited. This considered a reasonable assumption for non-breeder reactor cases, as there is always an excess of uranium in the system.

2.1.1.6 Waste Management

The waste streams tracked in the FIT calculations are the following: separations process wastes, fuel assembly cladding and structural materials, and quantities of the recycle fuel that is lost during fuel fabrication (in e.g. residuals that are held up in HEPA filters, containers, samples, gaseous discharges, waste water, scrap cladding, chemical solids, etc.).

Table 2.3 lists the various example waste forms used to immobilize respective separated waste isotopes from their respective separations process. Other waste forms could also considered for some of the waste streams, but addressing alternate waste forms is not pertinent for the objectives of this study. Comments on waste form choice and disposition possibilities are summarize in the lasts column of Table 2.3. [Piet 2012, Soelberg 2013]

Table 2.3: Separated isotope(s), process of origin, respective waste forms, and the respective rationale for the waste form choice. [adapted from Piet 2012]

Separated waste isotope(s)	From these separations processes	Waste form	Comments
C-14	All but electrochemical	Grouted carbonate (C-14 Grout)	Based on longevity and potential mobility, thus high probability for ingestion uptake pathway. (If the C14 is released to the atmosphere then there would be no C-14 waste form.)
I-129 (and co-collected halogens)	All	I-129 Glass Ceramic	Based on longevity and potential mobility.
Tc-99, UDS	UREX+1a	Metal Alloy Ingot	Based on longevity and potential mobility. Could be combined with HLW glass.
Tritium	All	Grouted Tritiated water (H-3 Grout)	Generally accepted disposition, although disposition as HLW may still be required; or free release may be allowed after sufficient decay prior to separations.
Kr-85 (and other Kr and Xe isotopes)	All	Compressed gas (Kr/Xe in cylinders)	Presumed disposition is disposal as LLW if allowed. Or store and then recycle or free release after sufficient decay time.
Cladding, UDS, baskets	Electrochemical	Metal Alloy Ingot (Echem)	Tc-99, other transition metals (except for some Zr), and other semi-metals including Te stay undissolved and partition in UDS with the cladding.
Raffinate/residual	All but electrochemical	Glass	Could include Tc, UDS, Cs/Sr, and lanthanides.
Raffinate/residual (salt waste)	Electrochemical	Glass-bonded zeolite (Echem)	Same as non-Echem raffinate/residual waste.
Cs/Sr	UREX+1a	Mineralized monolith	Could (or may be required to) be combined with HLW glass, depending on waste policy interpretation or economics.
Lanthanides	UREX+1a	Glass	Could be combined with HLW glass.
Cladding/coatings	Aqueous, Melt Refining, and AirOx	Compacted	Presumes residual TRU contamination >10 nCi/g.
Structure/hardware	All	Compacted (Remainder Metal)	Presumes combined with cladding/coatings.
Spent solvents-liquids (not yet in model)	UREX+1a	Mineralized monolith	Presumes TRU can be reduced to <10 nCi/g.
Fuel fabrication wastes	---	Compact	
Operations and maintenance wastes (not yet in model)	All	Mineralized monolith (combustible wastes only, after combustion)	Presumes some wastes contaminated with TRU, either < or >10 nCi/g.

A detailed description of all waste forms and detailed assumptions made for each waste form (i.e. parameters such as chemical makeup, void space, volume, etc.) can be found in the FIT User Guide [Piet 2012].

The user has the option to change the waste loading for some waste forms where appropriate (I-129 glass ceramic, metal alloy ingot, remainder metal, H-3 grout, C-14 grout); this is further explored in a parametric study (Section 3.3).

2.2 FIT Model Limitations

The FIT model is an effective tool for represents the processes in nuclear fuel cycles and their associated losses. However, in the context of this waste classification study, for which FIT was not originally designed, there exist many limitations that would need to be addressed in future work, if FIT is meant to become flexible enough to accommodate performing more thorough analyses of the back-end of the fuel cycle.

2.2.1 No Th

The FIT model currently does not support a choice of a thorium fuel cycle. To implement this functionality, the depletion code MrTau would have to be modified to accommodate a thorium reactor. In the context advanced fuel cycles informing a potential waste classification system, it would be useful to understand how the wastes created from a thorium fuel cycle differ from those of a uranium fuel cycle.

2.2.2 Waste Classification

The default waste classification in the FIT model is based on current classification categories. FIT does not allow for different classification for waste streams on the basis of changing criteria or a changing waste stream composition. For now, the model is designed to enable the user to select waste treatment options (such as treatments, waste forms, waste loading). The user is also asked to classify the waste streams manually. If FIT is to be used as a tool for better informing waste classification, future versions of FIT would ideally incorporate flexible and “smart” classification options for its waste streams; i.e. the manual classification should be replaced with one that asks users for desired criteria to judge waste classification, and have FIT automatically assign each waste stream to a class based on its composition and characteristics.

2.2.3 Limited Scope of Operation & Maintenance Waste

One severe limitation of FIT, from the waste management perspective, is the incompleteness of the inclusion of operational and maintenance (O&M) waste streams, some of which result in large volumes and mass, from fuel cycle processes into the model. The decision to leave these wastes out of the scope of the work at this time was made to enable the initial development of this systems model. No O&M waste from front-end processes, the initial fuel fabrication, or the two reactors in the cycle (the initial LWR and the recycle reactor) have been considered in the program implementation to date. However, O&M wastes for separations and fuel fabrication are included; FIT currently only deals with "technology wastes" that specifically contain wastes separated from the used fuel. So, the model includes tracking O&M waste masses for e.g. cladding and assembly materials. It does *not* account for contaminated materials from initial LWR fuel fabrication, contaminated equipment and low-level wastes from reactor operation, used fuel storage and transport, contamination HEPA filters generated during separations or fuel fabrication, ion exchange resins, waste water, waste solvents, spent equipment, PPE, etc.

AREVA and Energy Solutions recently published DOE Task Order 9 [AREVA 2013], "Improving the Estimates of Waste from Recycling", which provides an industry-based estimates for HLW and LLW from recycling. As of yet, these estimates have not been incorporated into the FIT model.

2.2.4 Missing Waste Streams

Also not included in the general FIT waste stream collection are upstream wastes such as uranium mine wastes and mill tailings and depleted uranium from enrichment. Any holistic consideration of a nuclear fuel cycle should take into account all wastes produced in every part of the fuel cycle. Mill tailings and DU present a non-negligible volume and long-term radiotoxicity problem.

2.2.5 Future Work for FIT Model Development

Despite these limitations of the FIT model in the context of the waste classification study, the waste stream analysis could still be performed within the bounds of the model. As described in Section 2.2.2, this study could serve as a starting point to develop FIT's "smart classification" capability. Furthermore, operational and maintenance wastes need to be included in future advanced fuel cycle waste classification work. The work performed in this study indicates how the scope of the FIT model could be expanded to serve as a tool to inform potential waste classification schemes.

Chapter 3

Results and Analysis: Heat as a Metric for Waste Classification

In order to understand the FIT model as a tool to inform waste classification of potential wastes generated from advanced fuel cycles, this chapter presents waste output metrics produced by fuel cycle simulations from FIT. It is discussed how these metrics can be used to inform a new approach to classifying nuclear waste according to its characteristics. The following sections describe the fuel cycle cases considered, the output metrics of choice, the results of a heat generation study and parametric study, and pertinent questions and how these results inform better waste management practices.

3.1 Selected Cases

Table 3.1 illustrates the set of reactor and separations choices in the FIT model that were utilized in this study. Indicated are the type of reactor and corresponding fuel and separations processes, and the age of the initial LWR UOX feedstock. The fast reactors of cases 1 and 2 can have different conversion ratios. The fast reactor fuel cycles discussed in Section 3.2 are evaluated using a conversion ratio of 0.5, and the sensitivity of varying this conversion ratio is explored in Section 3.3.

Table 3.1: The four representative fuel cycle cases considered.

Case	Separation 1	Reactor	Feedstock age	Separation 2	Cycles
1	UREX+1a	metal fast reactor	5 yrs	Electrochemical	19
2	UREX+1a	oxide fast reactor	5 yrs	UREX+1a	
3	PUREX	MOX	5 yrs	NA	1
4	PUREX	MOX	50 yrs	NA	

These cases represent a standard sample set of possible fuel cycles, within the limitations of the FIT model, which are alternative to the current open fuel cycle in the United States. Cases 1 and 2 represent two versions of a closed fuel cycle with 19 recycle loops (the first recycle of LWR UOX spent fuel plus 18 recycles after the first pass in the recycle reactor, totaling a maximum of 19 cycles, as limited by the FIT model). Cases 3 and 4 represent a modified open fuel cycle with one recycle into a MOX reactor, similar to the current French fuel cycle. For these MOX cases, two different feedstock cooling times—5 years and 50 years—were compared. The 50-year-old feedstock case represents the option of processing legacy used fuel, and explores the impacts of decades-long cooling of used fuel. Where applicable in this chapter, the once-through LWR fuel cycle has been labeled as "case 0" for comparison to the other cases. While additional fuel cycle options are possible, this set of fuel cycle options enables a representative exploration of the impact of fuel cycles on nuclear waste management and waste classification.

The fuel cycle options cases that were chosen in this study represent a robust range of fuel cycle options under consideration in the United States. Firstly, the reactor choices of each representative case are consistent with the DOE Fuel Cycle Research & Development program's three fuel cycle strategies of once-through, modified open, and full recycling [DOE FCRD]. Furthermore, the choices of reprocessing technologies used in FIT represent the a range separations options of varying complexities and types: a simple aqueous separations process (PUREX), a more complex multi-step aqueous process with various resulting waste streams (UREX+1a), and a high-temperature pyroprocessing technique using molten salts and metals. Adding another separations technology to the set of options, such as COEX or NUEX, as discussed in Section 2.1.1.2, may only marginally affect the results of this study, as other processes are similar derivatives of or variations on the processes chosen in this study.

3.2 Heat Generation

Because heat generation is generally the most important factor limiting geological repository areal loading and the capacity of repositories, the heat generation rate and its variation over time are key metrics of interest when attempting to make a first-order classification judgment. Since a geologic repository is ultimately necessary independent of fuel cycle choice, maximizing disposal capacity of the repository is a key issue. Disposal capacity directly correlates with heat load of emplaced waste packages; the peak near-field temperature determines the maximum heat loading of the waste package and the peak far-field peak temperature determines waste package spacing in the repository. The former is normally influenced by fission product heat load, and the latter is usually determined by heat-generating actinides. This section presents and discusses the FIT-generated heat rate values for each fuel cycle of choice, and discusses the impact of using heat rate as a metric for waste disposal options.

The units of the waste output metrics handled in the FIT model are expressed on a per-year basis (e.g. W/yr for heat generation rate). This unit is based on the amount of initial heavy metal (iHM) in the system, i.e. the incoming LWR-UOX feed, which is expressed in the units of kg iHM/yr. However, values and graphs in this chapter have been normalized to amount of initial heavy metal (kg iHM) and electricity generation (MWe) of the fuel cycle instead. "Initial heavy metal" is the measure adopted in the 1982 U.S. Nuclear Waste Policy Act as a metric to limit total repository capacity, but it provides an imperfect measure for actual waste hazard and disposal cost. More direct measures, such as heat generation and long-term dose, are more appropriate when evaluating repository capacity or performance. To be able to evaluate the efficacy of a fuel cycle choice as a whole, all unit values are normalized to the total amount of electricity (or proportionally, total thermal power) generated, with the simplified assumption that in the fast reactor cases, each cycle corresponds to the equilibrium values of the last cycle (of the set of 19 cycles). Waste output metrics are also expressed on a per-kg iHM basis to enable comparison of potential repository benefits based on current statutory limits.

3.2.1 Analysis Assumptions

No thermal model was developed to demonstrate the heat profile of each waste form (or package) in its host geology; yet, it is still possible to make a deduction about repository impact based on the heat generation of each waste form meant for a waste package bound for the repository. Short-term fission product heat can be dealt with by decay storage, so it may be advisable to classify wastes according to which materials would benefit from interim storage. It is important to point out that the heat generation of individual waste forms (i.e. waste forms before packaging for transport and disposal) are analyzed without assumptions about final package dimensions.

For the cases presented in Table 3.1, the short- and long-term heat dose contribution from each resulting waste stream was evaluated. For relevant heat-contributing isotopes, a Bateman solution was used to calculate decay and account for ingrowth of the isotope and its decay chain. This was done using a spreadsheet tool [Bays 2010, Piet 2011b] that implements the recursive solution to the Bateman equation [Shultis 2007]. It has been benchmarked against ORIGEN2.2 isotopic compositions after decay between 10^4 years and 10^9 years [Croff 1983].

In all figures representing each fuel cycle case, the cumulative heat generation rate at the point of the last recycle iteration is shown. For the fast reactor fuel cycle cases, this refers to the heat generation after 19 iterations, and for the MOX recycle cases, after just one pass of refabricated MOX fuel in the LWR recycle reactor. The total heat contribution of a multi-recycle system asymptotically approaches a limit as the reactor approaches equilibrium with each cycle. All heat generation values that are expressed in watts per kg of initial heavy metal (iHM) are based on the value of 19,532 kg iHM/yr of LWR-UOX fuel feed coming from a light-water reactor. This value is derived from an assumed burnup of 51 MWth-day/kg-iHM of the initial LWR, a 1 GWe (gigawatt-electric) power output, a capacity factor of 0.9, and a thermal efficiency of 0.33.

3.2.2 Waste Form Heat

Figures 3.1a and 3.1b show the total decay heat from all the waste forms resulting from each FIT-specified fuel cycle case as well as from LWR-UOX spent fuel. The two figures show the same data normalized to kg iHM and MWe, respectively. This is the sum of the heat contribution of all waste forms shown in Table 3.1. All waste streams generated in the FIT model "[result] from the reprocessing of spent nuclear fuel" [42 USC 10101] as described in Chapter 1; therefore, under the current legal framework, all waste streams should be classified as HLW. However, there are waste streams produced, most notably Cs/Sr, that may have the potential to significantly alleviate the short-term heat burden if separated, classified, managed, and disposed of differently.

The heat generation rate curves shown in Figures 3.1a and 3.1b are the sum of the heat contribution of all waste streams produced by each fuel cycle, as listed in Table 2.3. They do not show how many waste forms in total each fuel cycle produces; this depends on the amount of initial feedstock material.

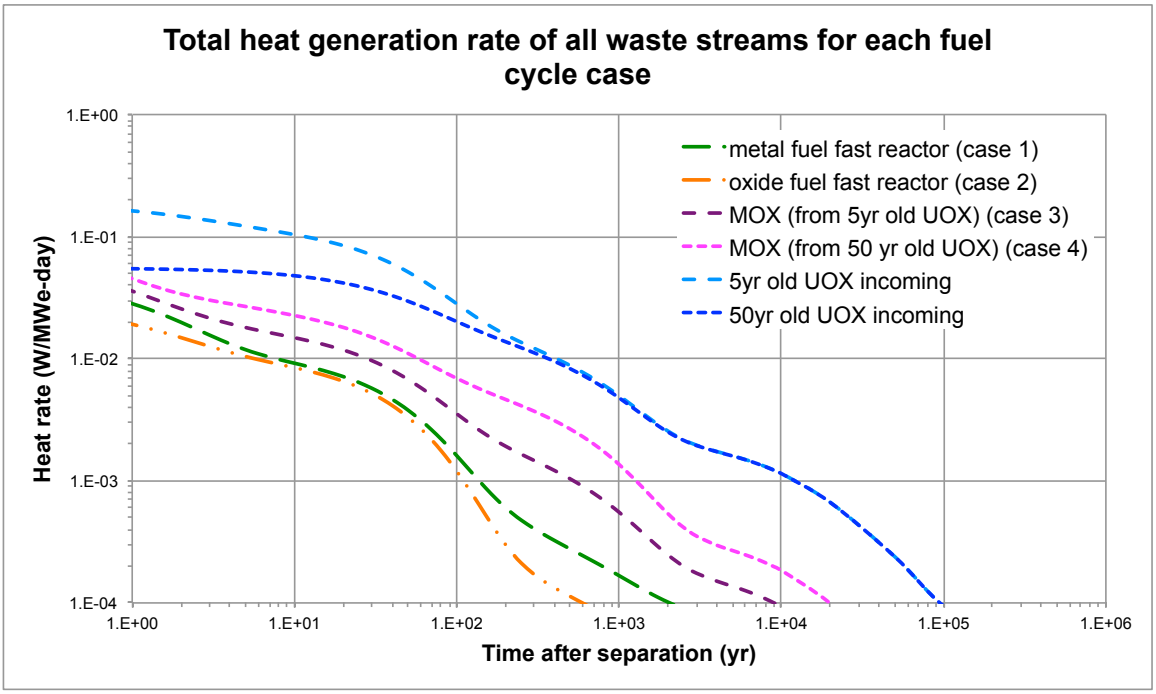

Figure 3.1a: Comparison of heat contribution from all waste forms of cases 1-4, including a base case comparison to two UOX spent fuel cases. Normalized to the total electricity generated for each fuel cycle.

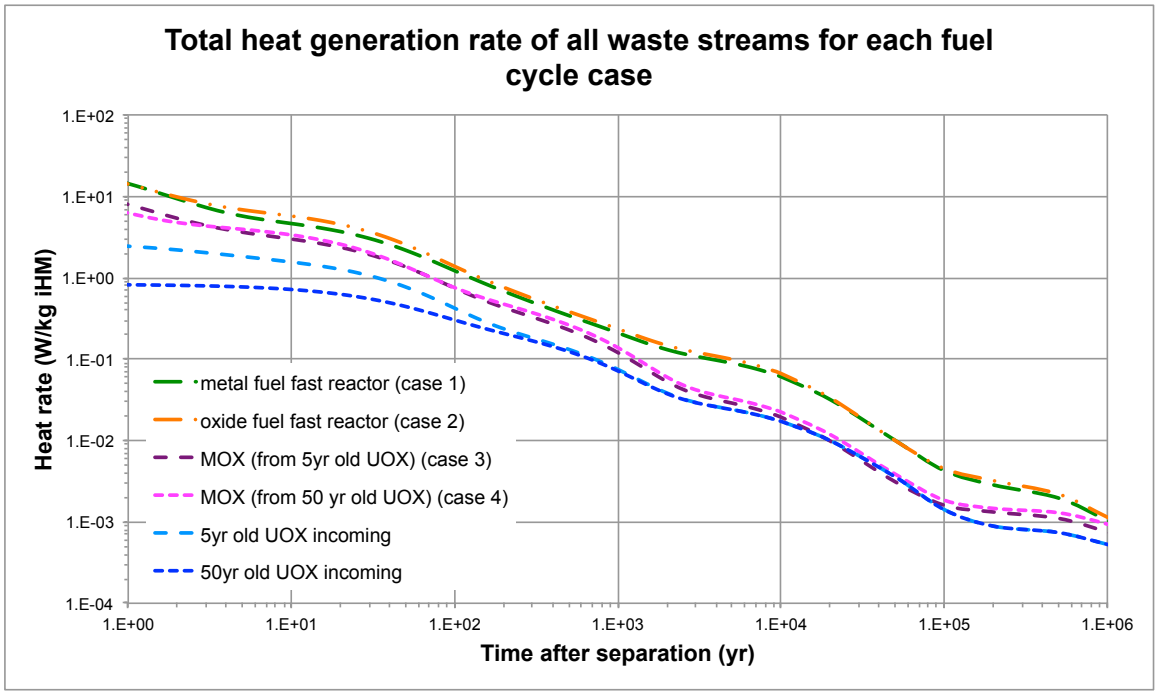

Figure 3.1b: Comparison of heat contribution from all waste forms of cases 1-4, including a base case comparison to two UOX spent fuel cases. Normalized to amount of initial heavy metal needed for each fuel cycle.

Figure 3.1a demonstrates how much total heat each fuel cycle contributes as normalized to total electricity produced for each fuel cycle choice, whereas Figure 3.1b shows the heat generation output per kg iHM. In Figure 3.1b, the two fast reactor cases are the greatest cumulative heat contributors, both in the short term and the long term. This is due to the 2-3 times larger burnup of fast reactors as compared to light-water reactors (see Table 3.2). However, due to their higher burnup and multiple recycles, the fast reactor fuel cycle cases generate a much larger cumulative amount of electricity per amount of waste heat produced and have better uranium utilization over the lifetime of the fuel cycle, as demonstrated in Figure 3.1a. Therefore, there exists a tradeoff of more waste heat for the benefit of larger electricity generation. Similarly, the spent fuel cases have the greatest total heat contribution as normalized to total electricity generation but the lowest when normalized to amount of initial fuel mass.

The values shown in Figure 3.1a (and all subsequent figures normalized to electricity generation) are based on the values shown in Table 3.2. Table 3.2 summarizes the burnup and electricity generation corresponding to each fuel cycle case. The electricity generation for each cycle was calculated by adding the electricity generated from the recycle reactor and the electricity generated from the number of initial LWRs needed to support the recycle reactor (which is calculated by FIT based on enrichment needed for the recycle reactor). Figure 3.1a is normalized to the total electricity generation of all 19 cycles. It is important to note that in both fast reactor cases, the total heat contribution from the unprocessed spent fuel is much less than that of other waste streams, since there is only one final unprocessed spent fuel bundle at the end of a multiple-recycle system. The total heat generation values of unprocessed spent fuel in the fast reactor cases have been normalized to the electricity generated by 19 cycles; however, the heat contribution of the unprocessed spent fuel would further decrease with each iteration. It is also important to note that Figures 3.2a—3.5a and 3.12a—3.14a represent heat generation values as normalized to electricity generation *per cycle*. This was done because comparing characteristics of individual waste forms is important in the classification analysis. It is nevertheless important to understand that the heat contribution of the unprocessed spent fuel bundle in fast reactor cases is large compared to other individual waste streams generated, but small if compared to the total waste generated over the lifetime of the fast reactor fuel cycle. For cases 1 and 2, unprocessed used fuel created per cycle gets fully reprocessed for the next pass in the reactor; the only unprocessed used fuel per set of fuel cycle recycles is at the very end of multiple recycles. Thus, if normalizing total heat contribution to the total electricity generation of the entire fuel cycle, the unprocessed used fuel heat generation curves only should be divided by a factor of 19.

The LWR-UOX spent fuel base cases are represented as cases 0a and 0b (aged 5 years and 50 years, respectively). Cases 1 and 2 are also divided into parts 1a, 1b, 2a and 2b, corresponding to a conversion ratio of 0.5 and 0.75 as shown in Table 3.2. A thermal efficiency of 0.44 was assumed for fast reactor cases and 0.33 for thermal reactor cases. A capacity factor of 0.9 was assumed for all reactors.

It is important to note that case 4, the MOX recycle from 50-yr-old feedstock case, shows an electricity generation of almost half of that of case 3, which originates from a required enrichment of ~4% (as opposed to ~9% for case 3), as calculated by the fuel input recipe of the reactor model in FIT. Since the Pu content of the spent fuel of case 4 should be lower than that of case 3 due to decay of Pu-241, this may be due to a mistake in the input fuel recipe, and will be addressed in future work.

Table 3.2 summarizes the quantities discussed above. The burnup values shown in Table 2.2 are slightly different because fuel with no impurities is assumed, whereas the burnup shown in Table 3.2 is based on an adjusted fuel recipe due to impurities in the fuel [Piet 2012]. The total electricity generated shown is on a per cycle basis. For the once-through and MOX cases (cases 0, 3, and 4), there is only one recycle loop. In fast reactor cases 1 and 2, the total electricity generation values shown are those of the last cycle, which correspond to the cycle that is closest to equilibrium composition of the reactor fuel.

Table 3.2: Burnup for each reactor for each fuel cycle case, and total electricity generation per cycle for each fuel cycle case. Two options of two different conversion ratios are shown for each fast reactor case. The fuel time residence in the recycle reactor does not vary significantly with multiple recycles. Adapted from [Piet 2012].

Case	Recycling reactor	Neutron spectrum	Burnup (MWth-day/kg iHM)	Thermal efficiency	Total electricity generation per cycle (MWe-day/kg iHM)
0a	5yr-old LWR UOX spent fuel	Thermal	51	0.33	15
0b	50yr-old LWR UOX spent fuel				
1a	FR metal, TRU, CR=0.50, from 5yr-old LWR UOX	Fast	139	0.44	202
2a	FR oxide, TRU, CR=0.50, from 5yr-old LWR UOX		182		206
1b	FR metal, TRU CR=0.75, from 5yr-old LWR UOX		102		170
2b	FR oxide, TRU CR=0.75, from 5yr-old LWR UOX		138		169
3	MOX-Pu, from 5yr-old LWR UOX	Thermal	45	0.33	133
4	MOX-Pu, from 50yr-old LWR UOX		47.7		70

Figures 3.2—3.5 illustrate a more detailed breakdown of heat-contributing waste streams for all fuel cycle cases. Generally, only the waste forms with the highest heat contribution are shown. This includes the last discharged core of unprocessed used fuel, of which there is only one over the entire set of cycles; as well as the Cs/Sr waste form (which in the metal fuel reactor case is only produced in the first recycle during aqueous reprocessing of the incoming spent fuel), and the glass-bonded zeolite (for case 1) or borosilicate glass (for case 2), which carry the bulk of the separated fission products.

In each of the subsequent figures, the heat contribution from fission products (FP) and actinides (U+TRU) in each waste form is shown. Despite partitioning efforts, actinides may be present alongside the fission products in the same waste form even if it is meant for just fission products. This is due to impurities and material losses during the separations processes; however, in certain waste streams they may be negligible. The "total" heat rate curve shown in each of these figures is the sum of all waste streams created per cycle. These waste streams (and their corresponding waste forms) are listed in Table 2.3.

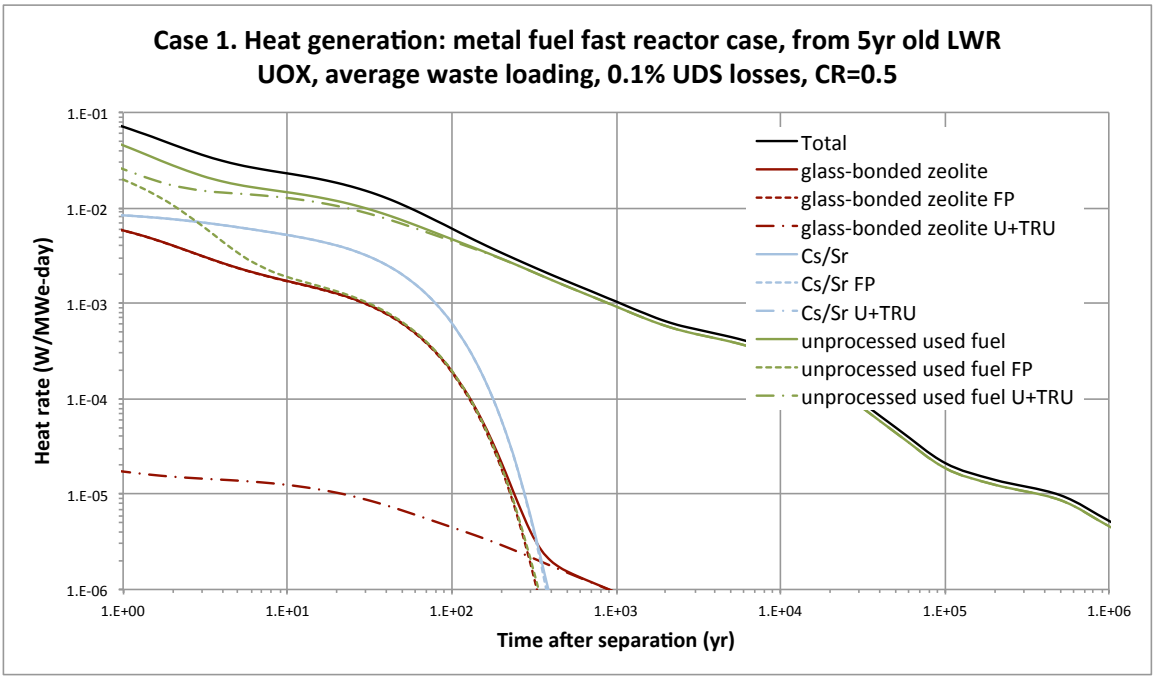

Figure 3.2a: Comparison of heat generation from Cs/Sr, glass-bonded zeolite, unprocessed used fuel waste streams for metal fuel fast reactor case (case 1). Normalized to electricity generation per cycle.

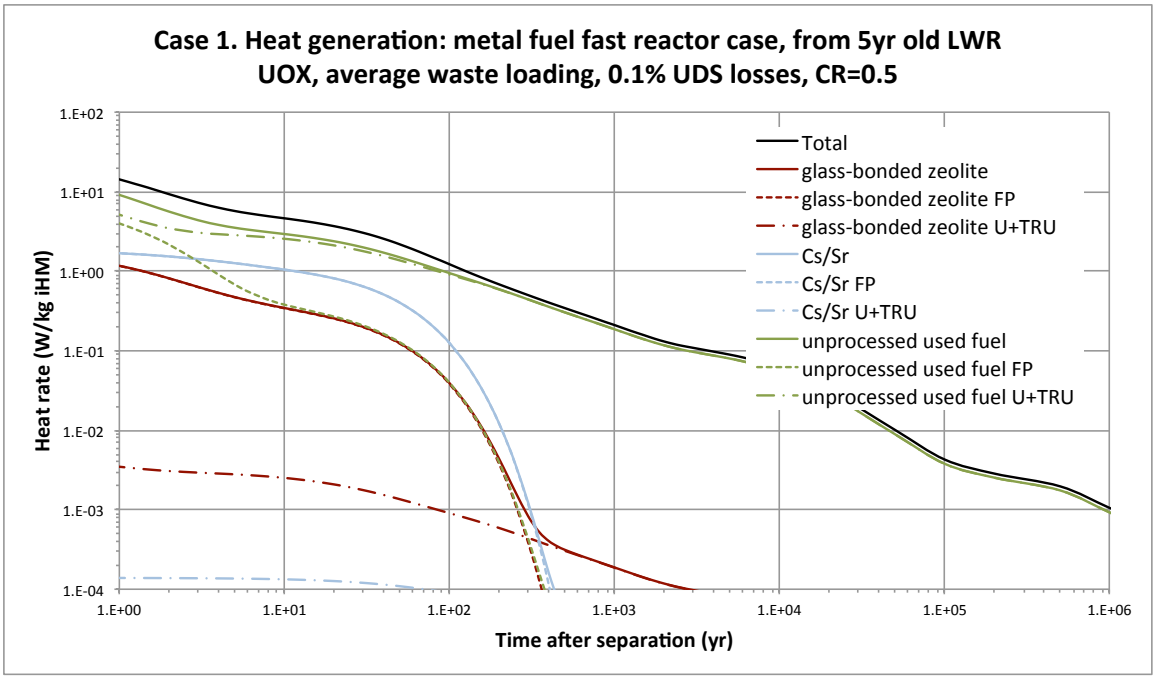

Figure 3.2b: Comparison of heat generation from Cs/Sr, glass-bonded zeolite, unprocessed used fuel waste streams for metal fuel fast reactor case (case 1). Normalized to amount of initial heavy metal needed from the initial LWR-UOX reactor per cycle.

Figures 3.2a and 3.2b show that the unprocessed used fuel bundle from the last recycle contributes the highest heat generation rate of the waste streams produced in one cycle. The glass-bonded zeolite and Cs/Sr waste forms are both relatively short-term heat contributors. For the unprocessed used fuel bundle, decay storage would offer little benefit to reducing the near-field peak temperature of the repository, as its heat burden is mostly generated by actinides. In contrast, the heat curves for the glass-bonded zeolite (the main waste form for fission products from electrochemical separation) and Cs/Sr (from the first UREX+1a separation) waste forms indicate that surface decay storage may be beneficial in terms of repository heat management. The main short-term heat load is carried by the isotopes Cs-137 and Sr-90, which decay below the heat dose level of the actinide impurities in the glass-bonded zeolite waste form after about 350 years and in the Cs/Sr waste form after about 450 years. After this timeframe, heat-generating actinides—mainly Am-241 from Pu-241 decay (with a half-life of 14.1 years) and Pu-238—drives the long-term far-field peak heat effects. Decay storage only makes sense for as long as the heat load of a waste form is dominated by short-term fission product heat, so the question thus becomes whether waiting for 350—450 years for fission products to decay is a reasonable timeframe for surface storage.

The separated Cs/Sr was calculated to decay to LLW Class C levels after about 320 years, applying the concentration limit rules of 10 CFR 61 for Pu-241 (Table I), Cs-137 and Sr-90 (Table II) concentrations [10 CFR 61.55].

Figures 3.3a and 3.3b tell a similar story for the oxide fuel fast reactor case (case 2). In this case, all materials are separated using the UREX+1a process and immobilized in appropriate waste forms as shown in Table 2.3. The heat generation rate of the three waste forms with highest heat load (HLW glass, Cs/Sr, and the unprocessed used fuel bundle) is shown. In this case, it was assumed that lanthanides were immobilized along with other fission products in the HLW borosilicate glass waste form.

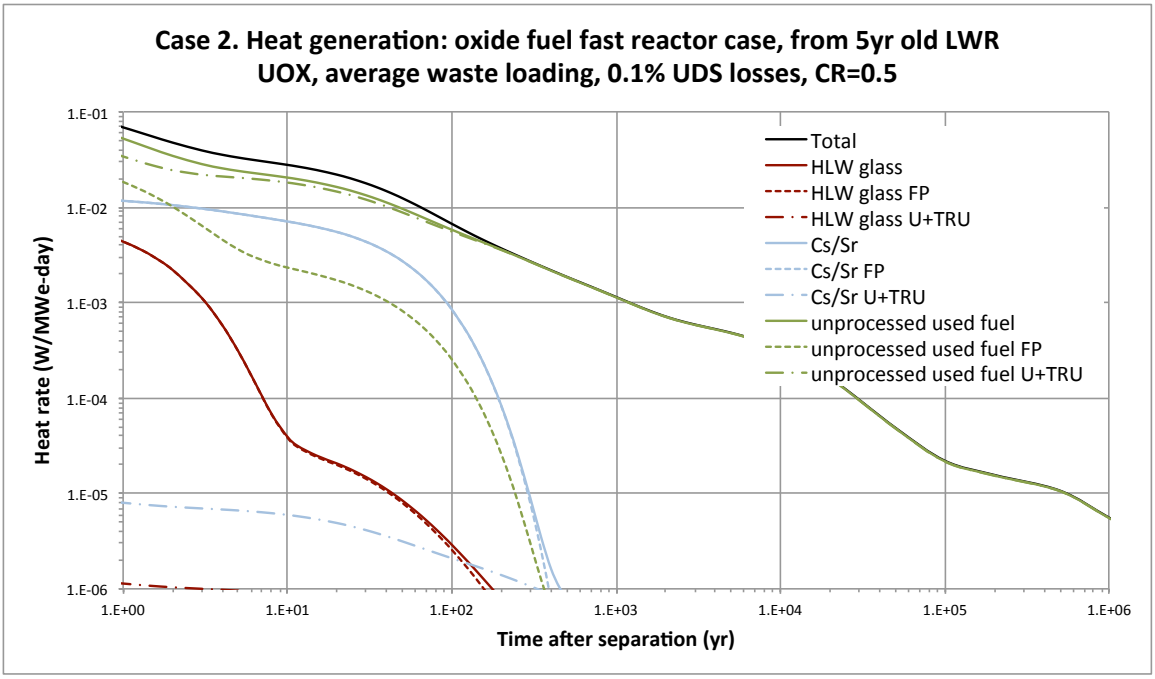

Figure 3.3a: Comparison of heat generation from Cs/Sr, HLW glass, unprocessed used fuel waste streams for oxide fuel fast reactor case (case 2). Normalized to electricity generation per cycle.

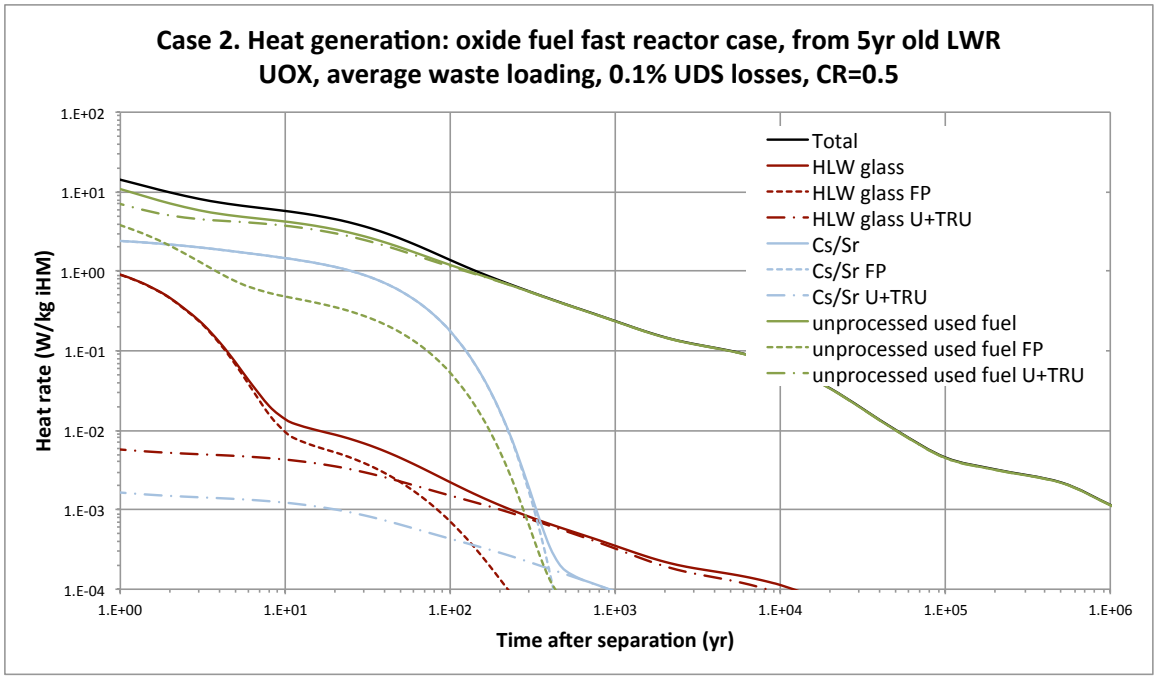

Figure 3.3b: Comparison of heat generation from Cs/Sr, HLW glass, unprocessed used fuel waste streams for oxide fuel fast reactor case (case 2). Normalized to amount of initial heavy metal needed from the initial LWR-UOX reactor per cycle.

The heat decay profile of the oxide fuel fast reactor case in Figures 3.3a and b is similar to that of the metal fuel fast reactor case (Figures 3.2a and b), showing that the main heat contributing waste form is the unprocessed spent fuel bundle discharged at the end of multiple recycles. In the Cs/Sr waste form, the fission product heat load falls below that of the actinides in the same waste form after about 350 years. In the case of HLW glass, this time point happens at about 50 years; thus, decay storage of several decades of HLW glass waste forms in this oxide fast reactor case may be an acceptable way of reducing the short-term heat burden and thus the near-field peak heat effects on the repository. Further purifying either of these waste streams by increasing the actinide recovery fraction could make long-term heat management of the repository even easier [Wigeland 2006]. As in the previous case, the separated Cs/Sr was found to decay to LLW Class C levels after about 320 years [10 CFR 61.55], after which near-surface burial would be acceptable.

Figures 3.4a, 3.4b, 3.5a and 3.5b show the heat load carried by wastes produced from the two MOX recycle cases.

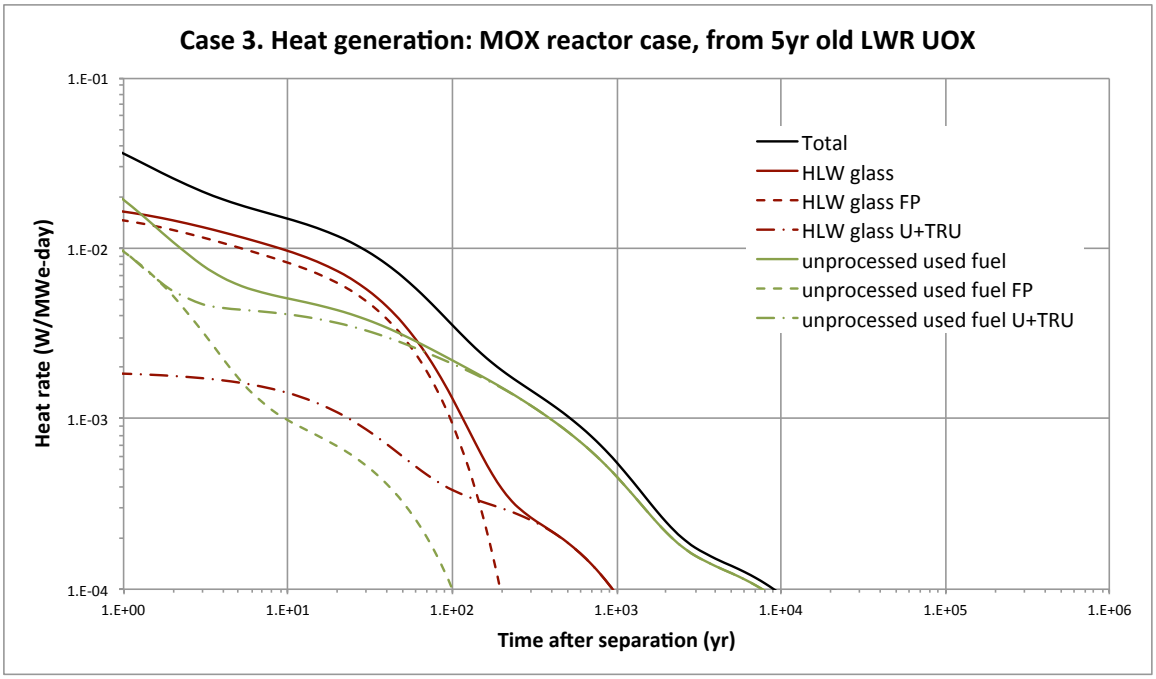

Figure 3.4a: Comparison of heat generation from HLW glass and unprocessed used fuel for 5-year-old feedstock MOX recycle case (case 3). Normalized to electricity generation per cycle.

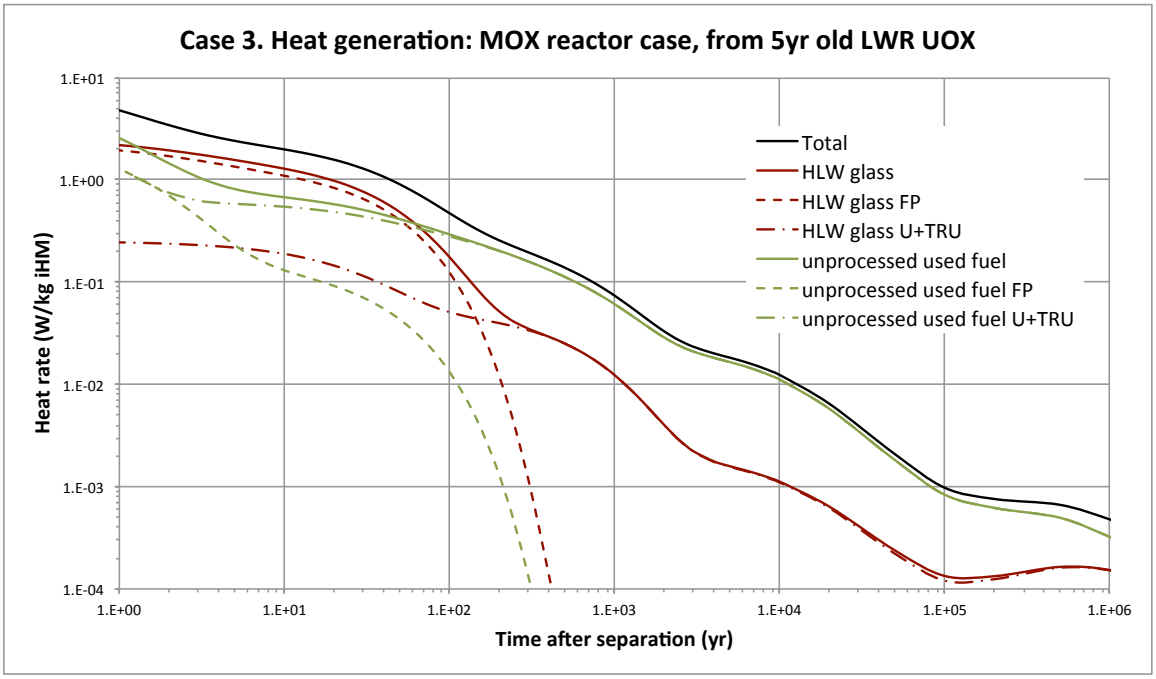

Figure 3.4b: Comparison of heat generation from HLW glass and unprocessed used fuel for 5-year-old feedstock MOX recycle case (case 3). Normalized to amount of initial heavy metal needed from the initial LWR-UOX reactor per cycle.

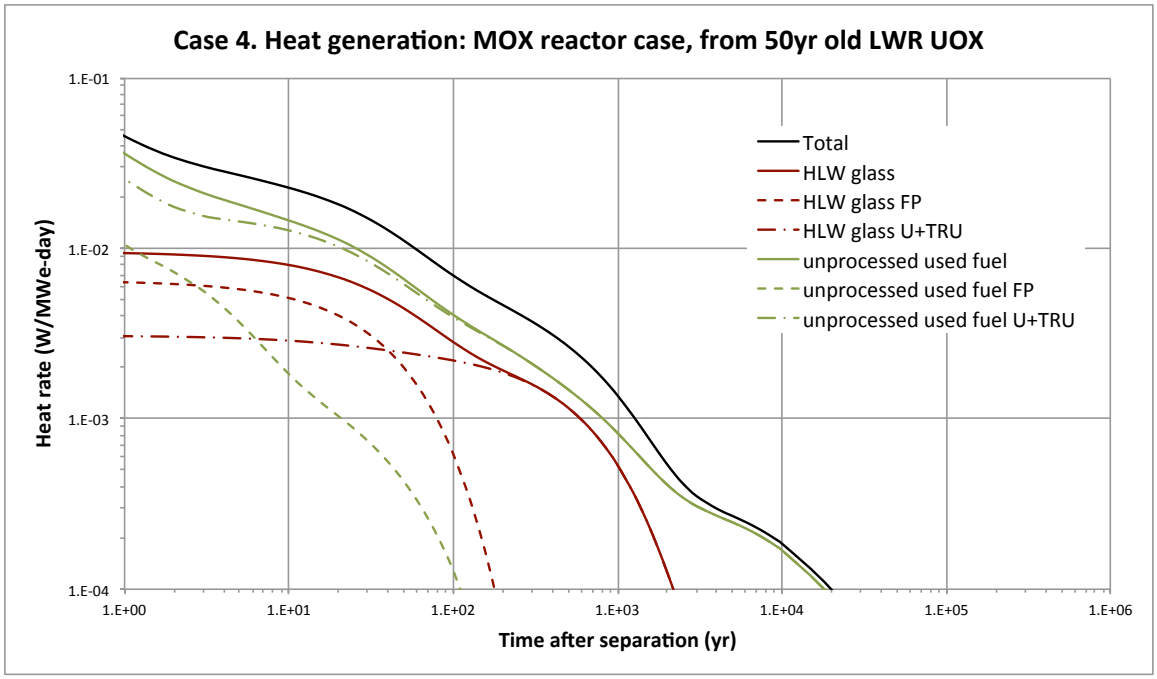

Figure 3.5a: Comparison of heat generation from HLW glass and unprocessed used fuel for 50-year-old feedstock MOX recycle case (case 4). Normalized to electricity generation per cycle.

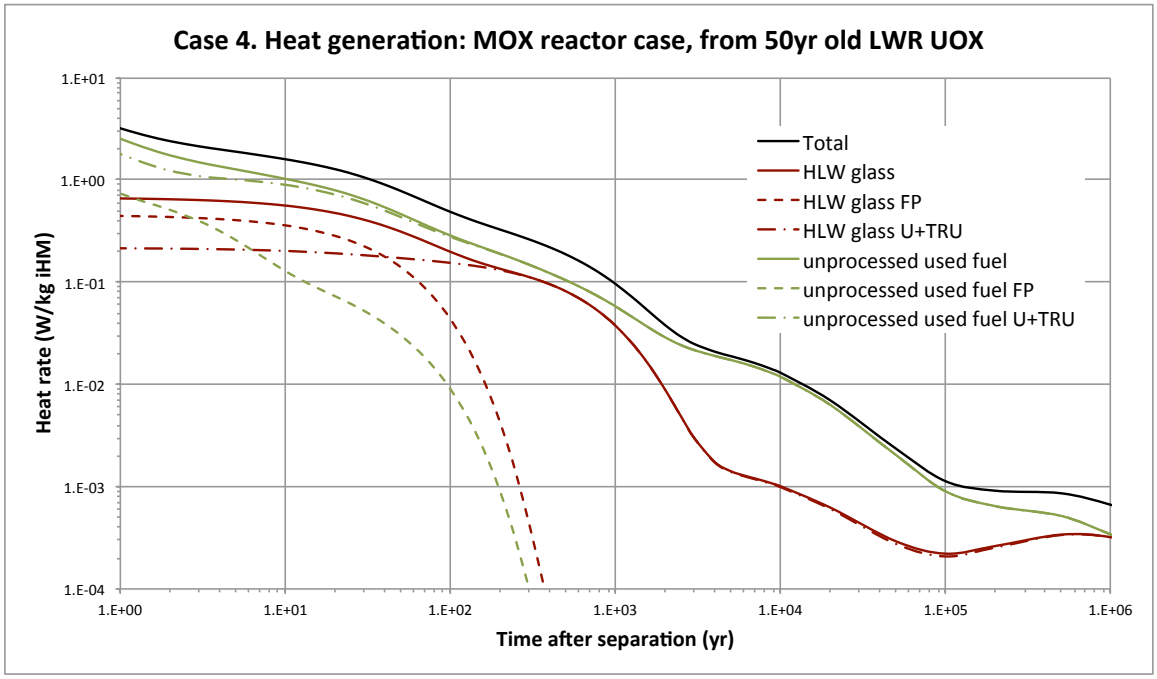

Figure 3.5b: Comparison of heat generation from HLW glass and unprocessed used fuel for 50-year-old feedstock MOX recycle case (case 4). Normalized to amount of initial heavy metal needed from the initial LWR-UOX reactor per cycle.

Figures 3.4a—3.5b show that the total heat load of both HLW glass and unprocessed used fuel for MOX-recycle cases are comparable. For the 5-year-old feedstock case, the heat generation from fission products dominates for the first 125 years until the actinide heat contribution takes over; for the 50-year-old feedstock case, the heat load is dominated by actinides (in both waste forms) from the beginning. In both cases, the total heat generation rate drops in half after about 450 years, which is when all the Cs and Sr have decayed to negligible levels and approximately half of the original Pu-241 and Am-241 have decayed. Whether there is a benefit to Cs/Sr separation for a MOX case is questionable; the idea of a 450-year-long surface storage institution, since it would need to be institutionally monitored for a very long period of time, may not be acceptable. Separation of Cs/Sr only makes sense if a separations process was used where actinides need to be recovered for utilization in a reactor, as in cases 1 and 2. At least, it would be beneficial to store the high-level wastes resulting from a MOX recycle case above ground for some reasonable temporary period to reduce the heat load before emplacement in the repository.

3.2.3 Implications of Fuel Cycle Choice on Repository Management

Under the current radioactive waste classification framework, all separated materials from advanced fuel cycles are classified as high-level waste, therefore neglecting benefits of separation from the waste management perspective. However, some of these separated wastes could benefit from interim storage to increase repository capacity (and thus reduce repository cost), and changing the class of these wastes appropriately would better inform the heat management strategy of a waste disposition regime. The results of this study indicate the possibility to use repository capacity as a function of decay heat management as a metric for waste classification.

There are several possible management and disposal options for each of the HLW streams for each of the fuel cycle cases:

- Repository disposal of UOX spent fuel (open cycle) or MOX fuel (modified open cycle) without Cs/Sr separation, following some period of interim storage.

- Decay storage of separated heat-generating fission products (Cs/Sr), then LLW Class C disposition (or another class). This applies to cases where Cs/Sr is separated (fast reactor fuel cycles implementing UREX+1a separation), which in this discussion includes cases 1 and 2.

- Immediate repository disposal of separated heat-generating fission products (Cs/Sr). This would not violate the current legal framework of classification (provided the reprocessing ban is lifted). Repository cost could increase and capacity decrease due to additional heat load.

- Decay storage, then repository disposal of separated heat-generating fission products (Cs/Sr). This could potentially not violate the current legal framework of classification. Potential cost reduction of repository due to heat load reduction of HLW packages emplaced immediately (bypassing interim storage).

Interim decay storage for Cs/Sr (separated in the UREX+1a process) would result in either decaying to LLW levels after about three centuries (and go to a near-surface disposal if reclassified as such), or, in shorter decay storage timeframes, reducing the near-field peak temperature of the repository. This decision would depend on whether it is considered reasonable, socially stable, and cost-effective to operate surface storage facilities for 300 years. In the case of the HLW glass form from an oxide fuel fast reactor case, it seems sensible to decay the waste form for several decades (a more reasonable timeframe), then place the waste into repository disposal when the fission product-driven heat load falls below the heat dose contribution of the actinides.

If Cs/Sr separation is not an option, as in the MOX recycle cases, the results of this study indicate that some decay storage to let fission products decay to reduce the heat load is nevertheless beneficial for increased repository capacity. However, the high actinide heat load of MOX fuel means that it would take longer to decrease to heat rate levels similar to UOX spent fuel. Therefore risk, feasibility and cost would need to be considered for this period of time. Also, it was shown that the long-term heat burden is slightly smaller if fresher UOX feedstock is used to produce MOX, so from this perspective it is prudent to recycle UOX feedstock into MOX fuel as soon as possible in future fuel cycle scenarios.

It is important to point out that there will always be a need for a geologic repository even in the case of recycling and transmuting transuranics. A final used fuel bundle from the last reactor core as well as structural material losses and undissolved solids will always be a product of the fuel cycle and will need to be disposed of, no matter how many fast reactor recycle loops are performed.

Whether Cs/Sr is separated from the rest of the high-level waste stream of a given recycle scenario will directly influence the heat load of the resulting set of waste packages and thus affect the heat management strategy of the repository [Wigeland 2006]. In this discussion, it is assumed that Cs/Sr separation happens in the cases of fast reactor recycle (cases 1 and 2), but that Cs/Sr is not separated out of MOX spent fuel assemblies, nor from the initial LWR feedstock fuel (cases 3 and 4).

3.3 Parametric Study

To determine the sensitivity of FIT output metrics, the impact on heat load due to varying fuel cycle parameters was investigated. The FIT-specific fuel cycle parameters (besides the type of fuel cycle itself) that can be varied in the model are as follows: conversion ratio (for fast reactor cases), waste loading (minimum/average/maximum, for most waste forms where data is available), and the percentage of UREX+1a separations that go to undissolved solids (UDS) (when applicable, i.e. only when UREX+1a separations process is implemented). All parameters may affect the actinide contamination of different HLW streams; therefore exploring their impact on waste composition is of interest in evaluating the appropriateness of the FIT model as a tool for informing waste classification practices.

The values in Figures 3.6—3.10 are expressed in one normalized version only (output metrics normalized to kg iHM). Showing one version of these graphs is sufficient to demonstrate fuel cycle metric sensitivity to an output parameter.

3.3.1 UREX+1a Losses to UDS

Varying the UREX+1a loss percentage to UDS had an impact on waste composition only of the waste forms associated with the UREX+1a process. In the metal fuel fast reactor case, shown in Figure 3.6, the UREX+1a process is implemented only for the recycling of the initial spent fuel. Therefore, the heat contribution of the waste forms showing the loss of the initial spent fuel fraction from aqueous reprocessing in UDS could be negligible overall, though each individual non-Echem metal alloy ingot waste form is sensitive to the loss fraction. The total heat load of undissolved solids from pyroprocessing is dominant because the number of metal alloy ingot waste forms immobilizing the UDS (as well as cladding and baskets) is much higher than the metal alloy ingot associated with the UDS from aqueous reprocessing.

Although there are losses to structural material associated with the PUREX process as well, FIT does not have an option to calculate varying percentages of these losses for anything but UREX+1a. Therefore, because the MOX reactor cases do not implement UREX+1a reprocessing, it was not possible to evaluate variation of composition loss into structural material for the MOX reactor cases.

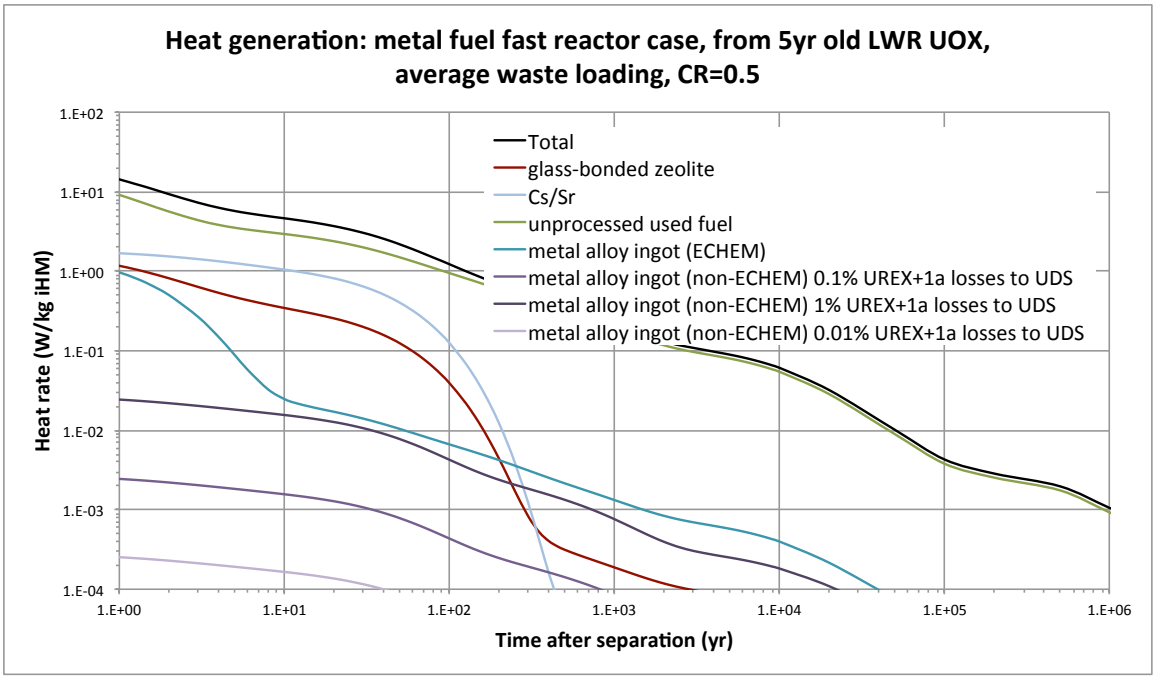

Figure 3.6: Comparison of heat generation from metal fuel fast reactor case (case1), varying the UREX+1a losses to UDS. Shown are metal alloy ingot waste forms from both UREX+1a and ECHEM reprocessing. Normalized to amount of initial heavy metal needed from the initial LWR-UOX reactor per cycle.

For the oxide fuel fast reactor case, it was shown that varying spent fuel losses in UREX+1a separations to UDS between 0.01%, 0.1%, and 1% had a proportional effect on waste form composition of the UDS immobilized in a metal alloy ingot. Varying the loss fraction by an order of magnitude affected the non-Echem metal alloy ingot waste form composition also by an order of magnitude, as shown in Figure 3.7.

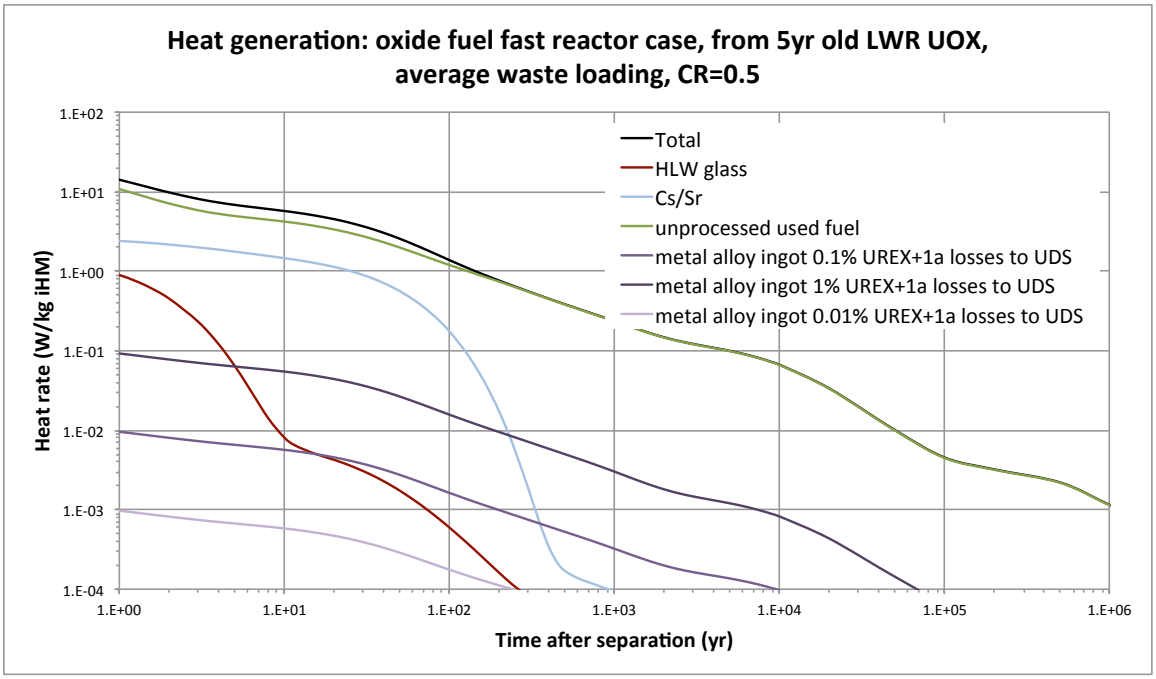

Figure 3.7: Comparison of heat generation from oxide fuel fast reactor case (case 2), varying the UREX+1a losses to UDS. Normalized to amount of initial heavy metal needed from the initial LWR-UOX reactor per cycle.

It is interesting to observe from Figures 3.6 and 3.7 that the composition of the metal alloy ingot from UREX+1a is proportional to the used fuel composition of the oxide fuel fast reactor, but the metal alloy ingot from ECHEM (and the metal fast reactor case) exhibits a sharp short-term heat load dropoff, presumably from fission product contamination from electrochemical separations process. It is also interesting to note that slight differences in the profile of the non-ECHEM metal alloy ingot arise. This is because waste compositions from different reactor equilibrium compositions from different iterations (e.g. 19th recycle of oxide fast reactor vs. 1st recycle of metal fast reactor) are being compared.

3.3.2 Conversion Ratio

Choosing a fuel cycle with a reactor of a different conversion ratio had a significant impact on waste composition. Conversion ratio is defined as the average number of fissile atoms created per fission event. In other terms, it is the ratio of the amount of fissile material produced in the reactor (and thus amount of fissile material discharged in the used nuclear fuel) to the amount of fissile material in the original fuel. In even simpler terms, it can be expressed as the TRU:U ratio. A conversion ratio higher than 1 means a breeder reactor; however, in the calculations run for this dissertation, all reactors were assumed to be burner reactors (CR<1). In these calculations, only two cases of burners with conversion ratios of 0.5 or 0.75, respectively, were used in the calculations. As a point of reference, current LWRs operate at a conversion ratio of about 0.6. [Nero 1979] Fuel from reactors approaching a conversion ratio of 1 must be reprocessed more often than ones with lower conversion ratio values, as the fuel can only be used for a shorter time to reduce the number of neutrons lost to absorption by neutron poisons.

A higher transuranic conversion ratio means more actinides produced. Figure 3.8 shows that the higher CR means higher heat generation for each set of waste forms except the Cs/Sr waste form—this is because the UREX+1a separation that is associated with creating this Cs/Sr waste form happens only during the first separation process (i.e. separation of the input LWR UOX fuel).

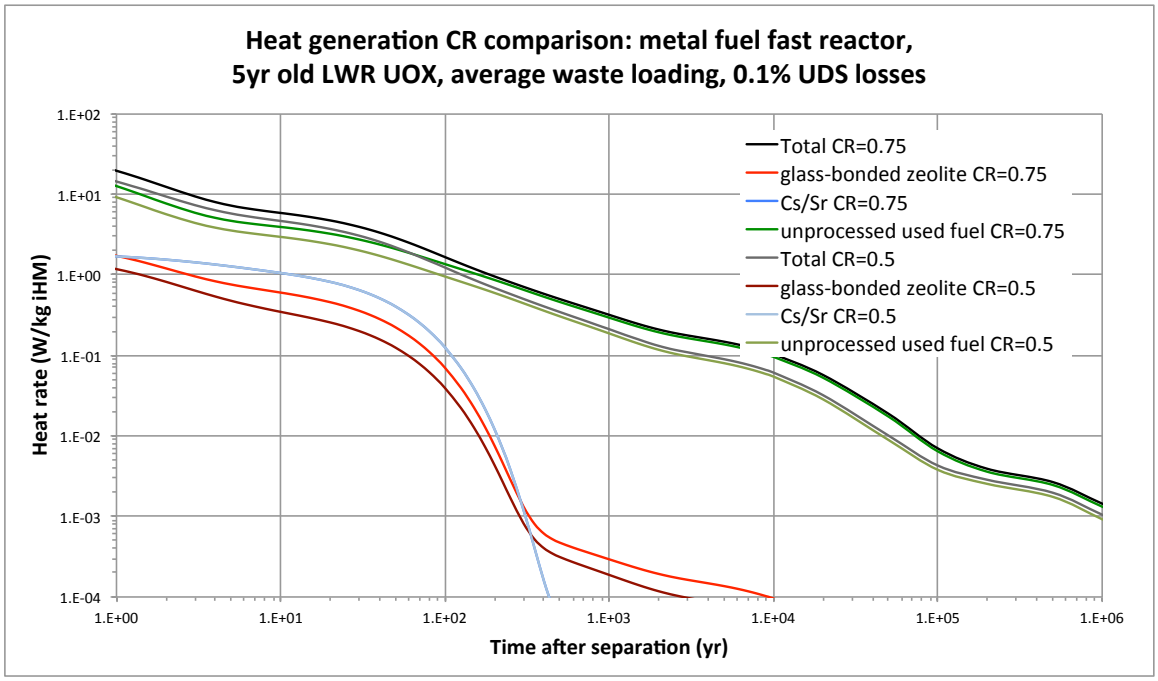

Figure 3.8: Comparison of heat generation from metal fuel fast reactor case, varying the conversion ratio. Normalized to amount of initial heavy metal needed from the initial LWR-UOX reactor per cycle.

Figure 3.9 shows the same trend of higher conversion ratio implying a proportionally higher heat content. In this figure, the heat content of the Cs/Sr waste form also increases due to the fact that the oxide reactor case separates Cs/Sr from the fast reactor oxide fuel in every recycle.

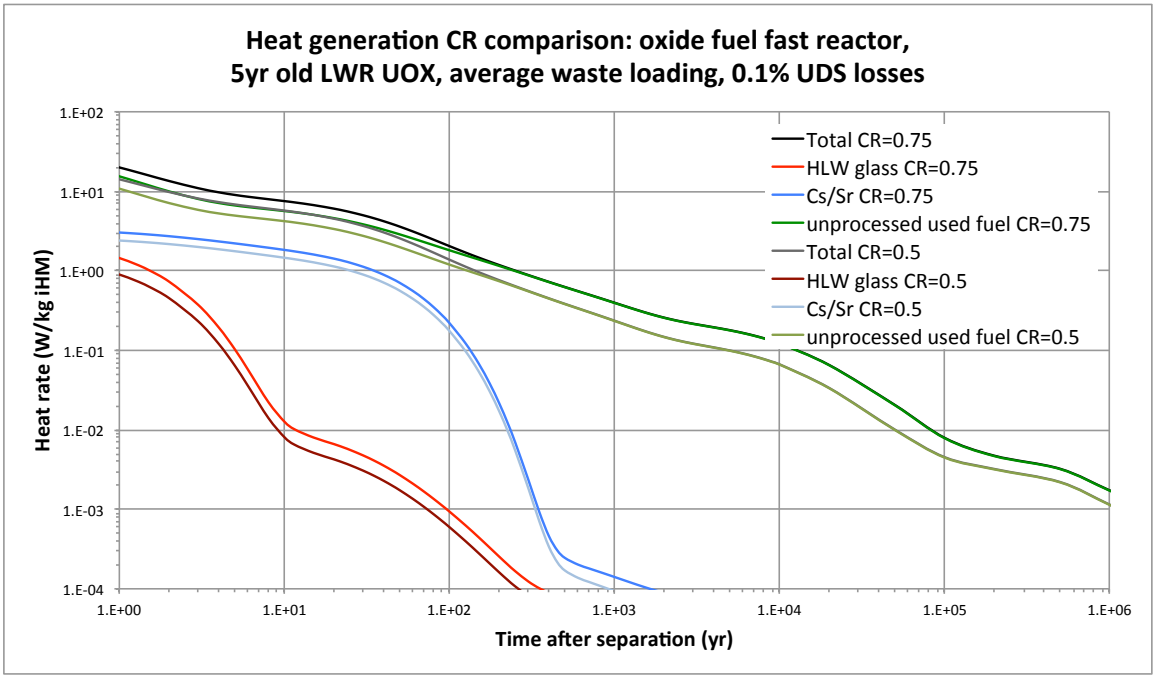

Figure 3.9: Comparison of heat generation from oxide fuel fast reactor case, varying the conversion ratio. Normalized to amount of initial heavy metal needed from the initial LWR-UOX reactor per cycle.

Figure 3.10 shows the heat rate as a function of recycle number of an example waste package (the unprocessed used fuel bundle, in this case). The figure demonstrates the effect that the conversion ratio value has on how many cycles it takes the fuel from the fast reactor to reach its equilibrium composition. It can be seen that the reactor with a lower conversion ratio (CR=0.5) reaches its equilibrium composition after fewer cycles than the one with the higher conversion ratio (CR=0.75). In other words, the higher the conversion ratio, the longer the reactor fuel composition and associated parameters (such as heat generation rate) take to reach equilibrium. Furthermore, the equilibrium fuel composition and heat rate are significantly higher for higher conversion ratios, due to higher burnups and therefore the creation of significantly more heat-generating fission products and transuranic elements in the waste stream. Therefore, increasing the conversion ratio also increases the heat burden of individual waste packages, but has the tradeoff benefit of higher electricity generation.

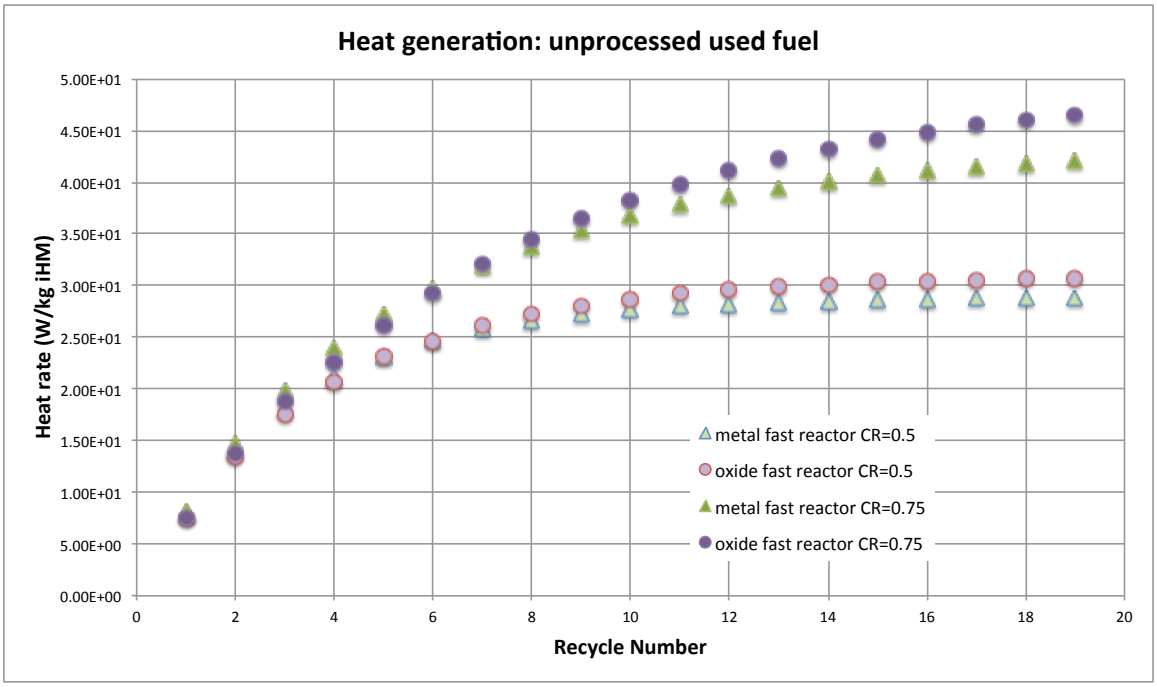

Figure 3.10: Comparison of unprocessed used fuel heat rate as a function of recycle number, varying fast reactor type and conversion ratio. (Example of a FIT waste parameter value of a waste stream as a function of recycle number.) Normalized to amount of initial heavy metal needed from the initial LWR-UOX reactor per cycle.

The results of the parametric study show that while heat generation rate is sensitive to varying parameters such as UDS loss percentage and conversion ratio, in any of these cases the total heat generation rate did not vary larger than a factor of 2. This is true even in the UDS losses case, where the metal alloy ingot carries an insignificant amount of the total heat burden; therefore, minimizing the amount of losses to UDS, to reduce the heat burden by one or two orders of magnitude, is beneficial only if this waste stream were treated separately from the rest.

3.4 Radiotoxicity

The concept of radiotoxicity is used as a measure of hazard, or informing another measure of hazard or risk. Therefore, radiotoxicity is an important metric for radioactive waste classification. However, the dose to humans from radioactive waste depends strongly on the waste form stability, engineered barriers, and the repository environment, which affect transport of and exposure to radionuclides. A repository model was not implemented in this study to evaluate the uptake pathway and dose rate of radionuclides to humans due to radionuclide migration; therefore it is not possible to quantify risk due to radiotoxicity from the waste streams. Using the measure of radiotoxicity of the waste composition alone would not be sufficient or accurate in informing waste classification with the aim of risk reduction of the waste. It would be misleading to "rank" individual radioisotopes according to their radiotoxicity, because the mitigation of risk by isolation of waste from the biosphere in the repository is not accounted for.

However, some general statements can be made about radiotoxicity and its usefulness as a metric. Long-lived, highly radiotoxic isotopes, such as I-129 and Np-235, are usually the most problematic radionuclides in long-term geologic performance assessment. The major difference in how radiotoxicity and heat are treated differently as waste classification metrics is that they both become important on different time scales. Heat is usually an issue on the order of decades to several centuries in geologic disposal cases, whereas radiotoxicity is an issue that extends into much longer timeframes, anywhere from the order of a thousand to a million years. [Piet 2013] This value also varies with geology.

Table 3.3 is based on [Nutt 2009, Clayton 2011, and Piet 2013] and summarizes recent studies done on generic repository environments and the relevance of radiotoxicity as a metric in each. Individual isotopes that were found to be dominant in transport (i.e. most mobile) through the geologic media are listed. Inventories of these radioisotopes can potentially serve as a metric for classifying wastes. All studies used UOX spent fuel as its source term for radionuclide transport.

Table 3.3: Summary of generic repository environments and dominant isotopes from respective performance study [adapted from Piet 2013].

Geology	Chemical environment (affects mobility)	Relevant isotopes
Tuff (generic environment)	oxidizing	Zr-93, Tc-99, I-129, Cs-135, U, Pu, Np, Am
Tuff (Yucca Mountain)	oxidizing	Tc-99, I-129, Ra-266, U, Pu, Np, Am
Salt	reducing (near-field), oxidizing (far-field)	C-14, Se-79, I-129, Pu, Np
Granite	reducing	I-129, Pu, Am
Clay	reducing	Cl-36, Se-79, I-129, Cs-135, Np, Pu, Am (C-14 and Tc-99 were relevant in another benchmark study)
Deep borehole	depends on zone	Cl-36, Tc-99, and I-129 (low doses)

Given the large variations in environmental parameters depending on specific geologies, it is difficult to derive conclusions about which generic environment is best in terms of performance and which radioisotopes are most hazardous in terms of radiotoxicity. This is true even if the same analysis had been done for wastes coming from advanced fuel cycles (i.e. all wastes discussed in this dissertation other than LWR UOX). According to [Piet 2013], "There are only modest differences among LWR-UOX, LWR-Th/^{233}U, and fast reactors for the relative importance of radiotoxicity among individual fission products," [Piet 2013] normalized to the number of fissions that took place (and therefore amount of electricity produced). However, actinide content could very widely. This is crucial because as Table 3.3 shows, actinides are important radionuclides in terms of dose contributors in 4 out of the 5 generic repository environment cases. One clear point is that a comprehensive waste disposal framework that has more than one type of repository could potentially allow much greater optimization of waste forms for disposal.

Ideally, a classification system should incorporate both metrics of radiotoxicity and heat generation to reflect long-term hazard of waste requiring permanent isolation and short-term difficulty of handling highly radioactive waste, respectively [OECD 2002, OECD 2006]. Table 3.4 shows a recent suggestion for a simple classification system [Bays 2010b]. The idea of partitioning waste up into the four boxes shown is to implement advanced fuel cycles to minimize the amount of waste in the "high heat—high long-term radiotoxicity" box, and by partitioning and transmutation, move the resulting waste streams into the other three boxes, the wastes of which are more manageable. However, the four categories this idea presents is not sufficient to handle some wastes that the IAEA classification guidelines recommends; for example, it does not present a category for exempt wastes nor does it distinguish between different types of short-lived waste, or between long-lived TRUs and fission products.

Table 3.4: Suggested radioactive waste characterization based on decay heat and radiotoxicity, and longevity of the radioisotopes in each category (adapted from Bays 2010b and Piet 2013).

	High Long-Term Radiotoxicity	Low Long-Term Radiotoxicity
High Heat	Example isotopes: Transuranics (and Trans-thorium, if applicable). Expected disposition: Deep geological disposal.	Example isotopes: ^{3}H, ^{85}Kr, ^{137}Cs, ^{90}Sr. Example disposition: de facto decay during storage of used fuel in the U.S. before final disposal.
Low Heat	Example isotopes: ^{14}C, ^{99}Tc, ^{129}I. Example disposition: Waste entombed in decommissioned U.S. nuclear facilities, German Konrad Mine, TRU waste in WIPP.	Example isotopes: Shorter lived fission products in low concentrations. Example disposition: Shallow land burial in the U.S. of waste that meets current Class A, B, or C LLW limits.

In a similar way, radiotoxicity as a metric could potentially be qualitatively coupled with that of heat [Soelberg 2009] to create a classification framework that is simple yet effective as the basis of a characteristics-based system. However, longevity, coupled with heat, might be a better measure to categorize waste classes due to the absence of a repository model (and thus no reasonable evaluation of disposal option effect on radiotoxicity hazard).

3.5 Classification

Whether choosing to close the fuel cycle with a continuous-recycle fast reactor system or recycling only once in a modified open cycle with a MOX reactor, advanced fuel cycles create waste streams that are not all in the scope of the current classification framework of the United States. Constructing a characteristics-based classification system is crucial to being able to treat all wastes created, regardless of the choice of fuel cycle. This also applies to the current open fuel cycle, which, as a result of an incomplete patchwork of classification rules, has led to the creation of wastes that fall under no legal framework.

The results discussed in this chapter indicate that waste heat is an important metric for informing not only general waste management practices, but also more specifically in waste classification. This section will explore the effects of classifying advanced fuel cycle-generated waste (as modeled in FIT) under the United States classification scheme, and will suggest a way to classify wastes according to waste stream characteristics, such that the creation of orphan wastes is eliminated. The goal is to suggest features of a "technology-neutral" classification system which are nevertheless informed by possible future fuel cycle scenarios as well as already-existing issues such as orphaned waste streams.

As this is a scoping study designed to study the relative impact of different fuel cycles, precise numerical values indicating waste class boundaries might not be too meaningful in this context. But, general guidelines are suggested, as informed by earlier findings of this study.

As previously mentioned, closing the fuel cycle would lead to the creation of many waste streams that would neglect the partitioning efforts from the waste management perspective. Therefore, it is crucial to implement a characteristics-based classification framework in order to categorize any type of waste potentially produced, regardless of its origin.

The idea of informing repository management based on disposal priority, as a function of process, waste form, and waste composition, is also discussed.

3.5.1 Classification of FIT-generated Waste under Current Legal Framework

In the FIT fuel cycle cases chosen for this study, many waste streams are created in the process of separation, fuel fabrication, and waste treatment. For the fast reactor cases, these new waste streams do not exist on a commercial scale in any country, but could become a reality if adoption of that fuel cycle were to become the case.

As mentioned, FIT does not yet treat operational and maintenance (O&M) wastes. These waste streams are crucial for making the case for an urgent need for a characteristics-based classification system, since their fate under the current classification framework is unclear. Future work on this subject should include the considerations of the recent AREVA/Energy Solutions Task Order 9 report on wastes incidental to reprocessing [AREVA 2013].

Classifying all waste "resulting from the reprocessing of spent nuclear fuel" [42 USC 10101] as high-level waste potentially neglects the waste management benefits that these fuel cycle options have to offer. The appropriate disposal levels and hazards attributes for varying resulting waste streams are quite different. One of the main values of classifying waste streams according to their differing characteristics lies in assigning appropriate credit to fuel cycle options for the reduction of the heat burden on the repository and the radiotoxicity burden on the environment by storing and/or disposing of different waste streams appropriately. Wastes generated in advanced fuel cycles can help inform how current regulations and policies could be revised to develop more efficient disposal concepts.

In this section, different FIT-generated waste forms are assigned current waste classes based on a correlation between current waste —classes and the characteristics of each waste stream. [Soelberg 2007] Figures 3.11—3.14 show, for each fuel cycle case, a suggested classification based on the current waste class nomenclature. Some assumptions may not be consistent with interpretations of current, source-based waste management policy and regulations. Under the current legal framework, all waste forms shown in Figures 3.11—3.14 should technically all be classified as HLW; except for the fuel fabrication waste stream, which is not a direct waste from the separations and treatment process of spent fuel, thus its definition being unclear. Although the existing waste classes are not perfectly applicable to these newly generated waste streams, these figures suggest a way to classify the different waste streams according to currently established classes of wastes as illustrated in Table 3.5, in an attempt to gain some insight on whether the current waste class definitions meet the needs of an advanced fuel cycle.

Table 3.5: Possible classification of each waste stream based on existing waste class definitions, and justification of assumptions [Piet 2012 and Soelberg 2013].

Waste	Suggested class	Comment
Iodine Glass Ceramic	HLW-Repository	Based on iodine's longevity and mobility. I-129 is a key radioisotope in many uptake pathway models that contributes to dose.
Kr-Xe	LLW-Decay Storage	Decays to low levels. Not mentioned in 10 CFR 61. Currently dealt with by atmospheric release.
Cs-Sr	MLLW-Decay Storage	Decays to low levels. Hazardous material, though potentially could meet modified LLW criteria.
Metal Alloy Ingot (Non-Echem)	HLW-Repository	Contains Tc. Applying source-based criteria; not necessarily justified.
Remainder Metal	LLW-GTCC	Cladding. Contains some FP and long-lived activation products.
HTO Grout	LLW	Short-lived. Tritium does not prevent a waste from qualifying as LLW-ABC.
C14 Grout	HLW-Repository	Based on C-14's longevity and high potential to contribute to uptake dose.
LLW Mineralized Monolith	LLW	By definition. Equivalent of solvents that are incinerated and put into waste form.
TRU Mineralized Monolith	LLW-GTCC	DOE-type TRU wastes tend to map into LLW-GTCC in NRC criteria.
Glass	HLW-Repository	Vitrified glass; contains most of the fission products from aqueous separation.
Glass-Bonded Zeolite	HLW-Repository	Fission product-bearing salt from electrochemical separation stabilized in this waste form.
Fuel Fab Waste	LLW-GTCC	Includes actinides (TRU). Low concentration of FP impurities.
Operational Waste	LLW	Essentially by definition, as there is a separate option for TRU-contaminated waste.
Operational Waste (TRU)	LLW-GTCC	DOE-type TRU wastes tend to map into LLW-GTCC in NRC criteria.
Lanthanide Glass	HLW-Repository	Combined with other FP in glass.
Metal Alloy Ingot (Echem)	HLW-Repository	Contains hulls and Tc.
Unprocessed Used Fuel	HLW-Repository	Spent fuel; HLW by definition.

The suggested waste classes for each of the waste streams from Table 3.5 are shown in the waste generation heat curves for each waste form in Figures 3.11—3.14. Again, both normalizations are shown in each version of the same figure, as indicated.

Figures 3.11a and 3.11b show the waste forms resulting from the metal fuel fast reactor case. All the waste forms that need to be classified as HLW in different shades of red. Some wastes with a low heat load, such as I-129 and C-14 (if immobilized, as opposed to being released into the atmosphere), are not shown. However, these could also need to be classified as HLW based on other metrics such as longevity and mobility. The Cs/Sr waste form has been classified as MLLW destined for decay storage. As discussed in Section 3.2.3, this may or may not be desirable based on repository management choices. Generally, using the heat metric to make decisions about repository management makes most sense for (high-activity) HLW instead of lower-activity wastes, but it is also useful in determining whether something could be classified as waste that is stored on the surface.

Another waste stream of interest is the fuel fabrication waste stream, which in composition is similar to TRU waste; it is long-lived and is contaminated by actinides. In Figures 3.11a—3.12b, it is suggested that it be classified as GTCC LLW due to its resemblance to DOE TRU waste but commercial (and not defense-related) origin. This example serves as an instance that underlines the ambiguity and arbitrariness of the current classification framework. It is also interesting to note that the metal alloy ingot, which is the waste form for UDS, has lower heat content than the fuel fabrication waste (after a short period of decay). In this example framework, it is classified as HLW, but only because it is a direct byproduct of the separations process and contains fission products; otherwise, this classification is arbitrary and this waste stream could probably qualify as LLW-GTCC.

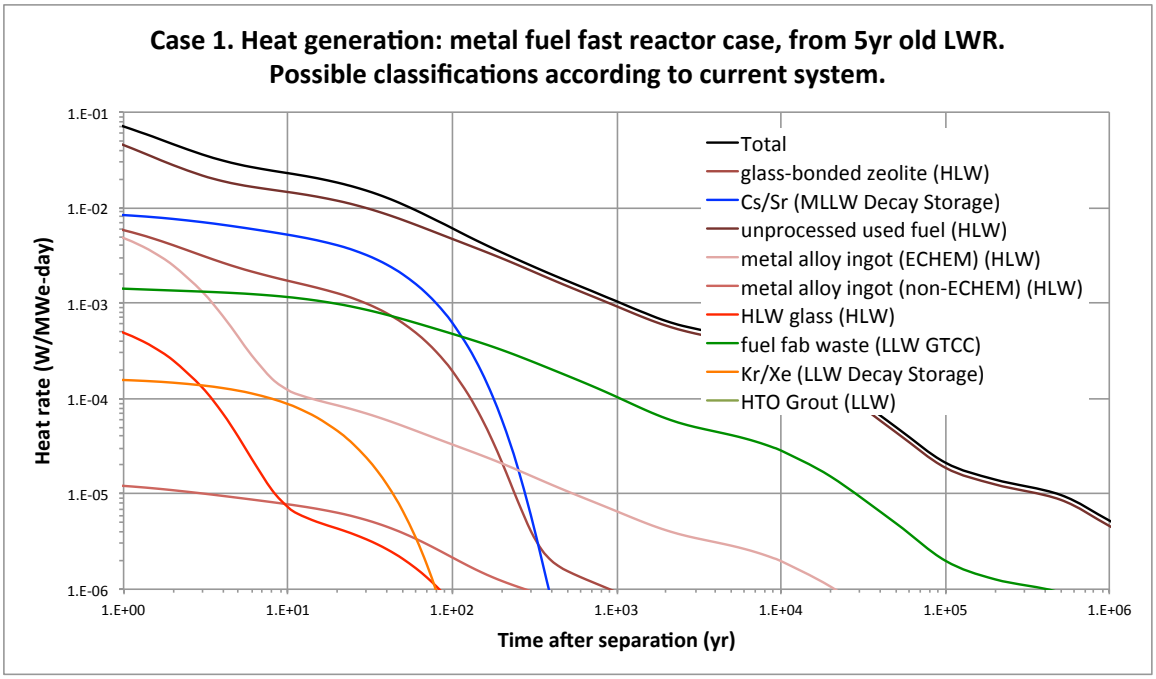

Figure 3.11a: Classification of all waste streams from metal fuel fast reactor case into existing classes. Normalized to electricity generation per cycle.

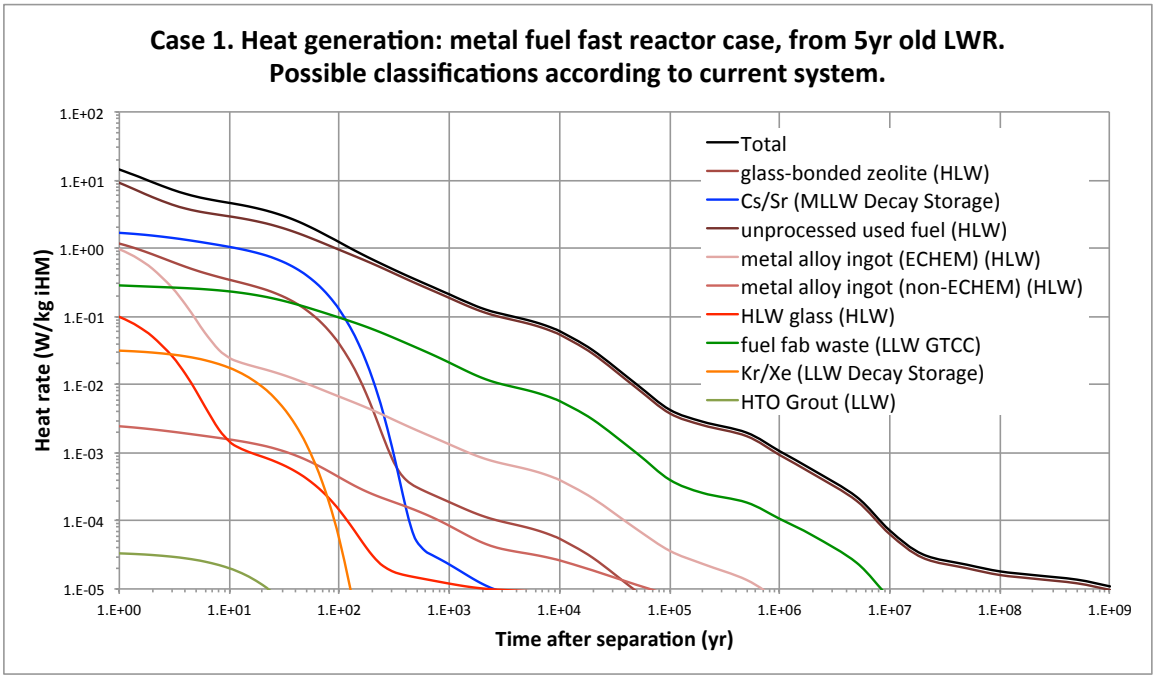

Figure 3.11b: Classification of all waste streams from metal fuel fast reactor case into existing classes. Normalized to amount of initial heavy metal needed from the initial LWR-UOX reactor per cycle.

Figures 3.12a and 3.12b show a similar attempt to categorize the oxide fuel fast reactor waste streams according to existing classes. Classified as high-level waste are vitrified glass, unprocessed used fuel, and metal alloy ingot (UDS), due to their high heat and high fission product content. The fuel fabrication waste stream has lower heat content than that of the metal reactor case but is likewise long-lived. Tritium waste (HTO grout) and the immobilized Kr-Xe waste form can be easily decayed to very low levels after at most 100 years, therefore some level of LLW class is applicable here.

For both case 1 and case 2 (Figure pairs 3.11 and 3.12), the Cs/Sr waste stream separated from the used fuel could be classified as MLLW ("mixed" LLW due to a nonzero chemical toxicity hazard), depending on long-term actinide impurities, and kept in decay storage until it decays to LLW GTCC levels, as discussed in Section 3.2.2.

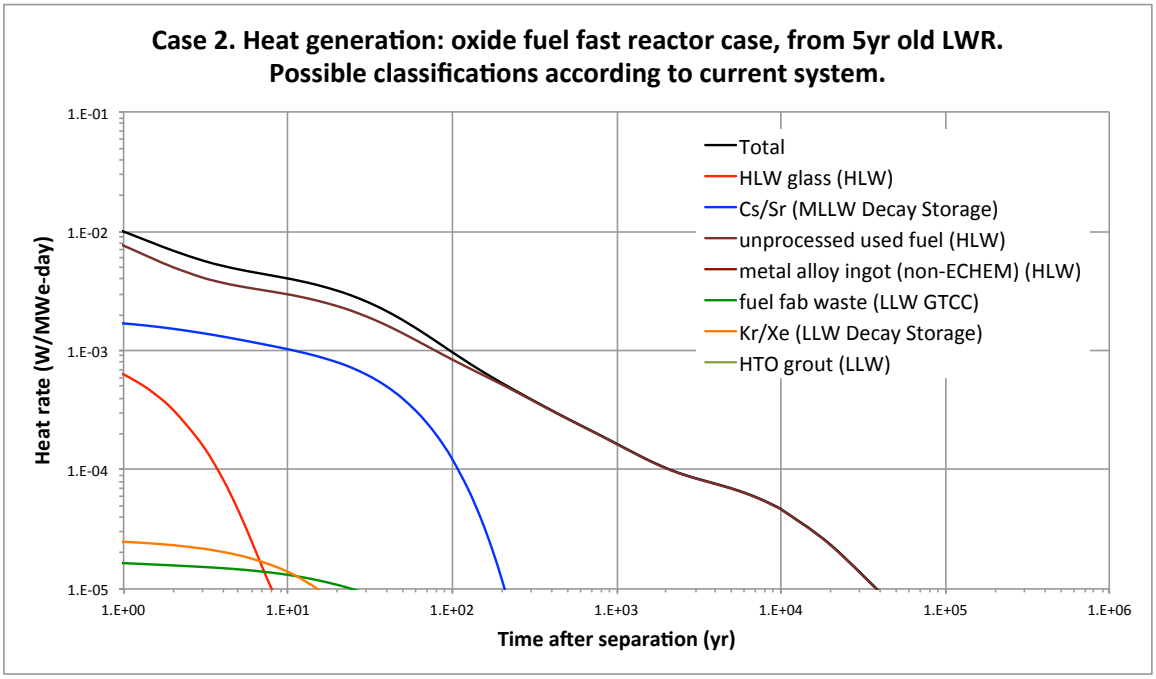

Figure 3.12a: Classification of all waste streams from oxide fuel fast reactor case into existing classes. Normalized to electricity generation per cycle.

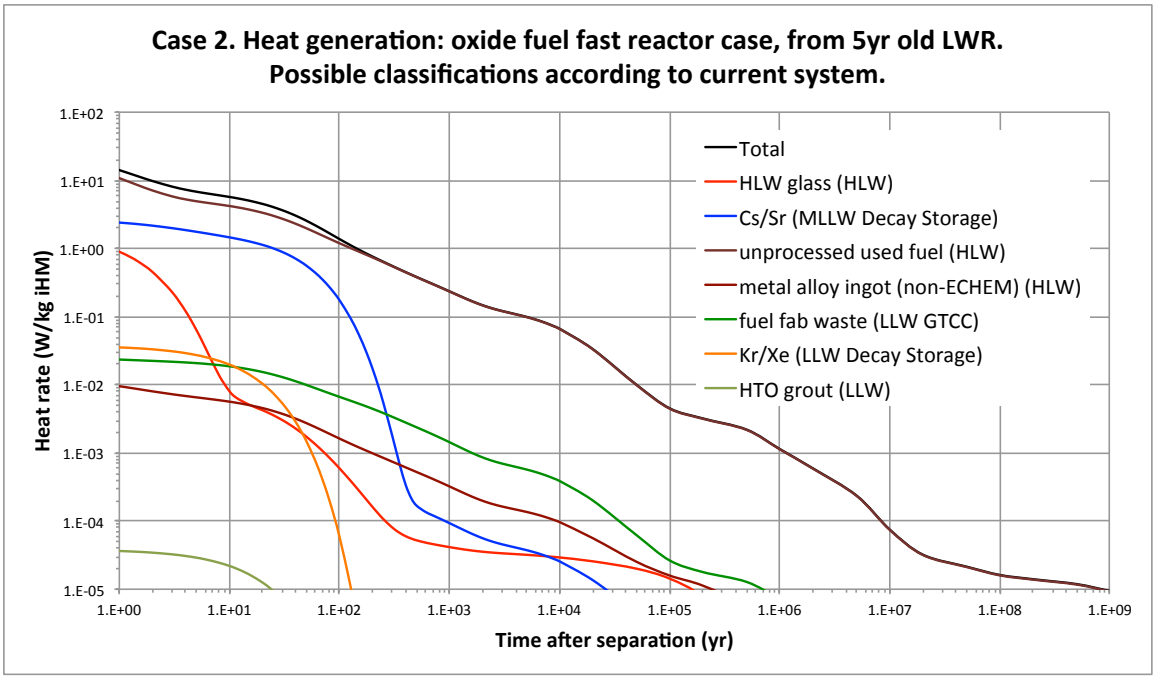

Figure 3.12b: Classification of all waste streams from oxide fuel fast reactor case into existing classes. Normalized to amount of initial heavy metal needed from the initial LWR-UOX reactor per cycle.

Figures 3.13a, 3.13b, 3.14a, and 3.14b show the possible classifications from the MOX reactor cases (cases 3 and 4, respectively). Both MOX cases are very similar to each other. In most waste form cases, the short term and long-term heat load for each waste stream is higher for the 50-yr old spent fuel MOX recycle case. The tritium waste stream in case 4 (the older MOX fuel) is not pictured because it has already decayed to very low levels. For the same reason, Kr/Xe is also present in much lower quantities in case 4. Also, the heat of the metal alloy ingot waste form becomes less than the heat contribution of the fuel fabrication waste stream at 20 years for case 3, and at 200 years for case 4. It could impact potential classification decisions if considering classifying the metal alloy ingot waste stream as something other than HLW.

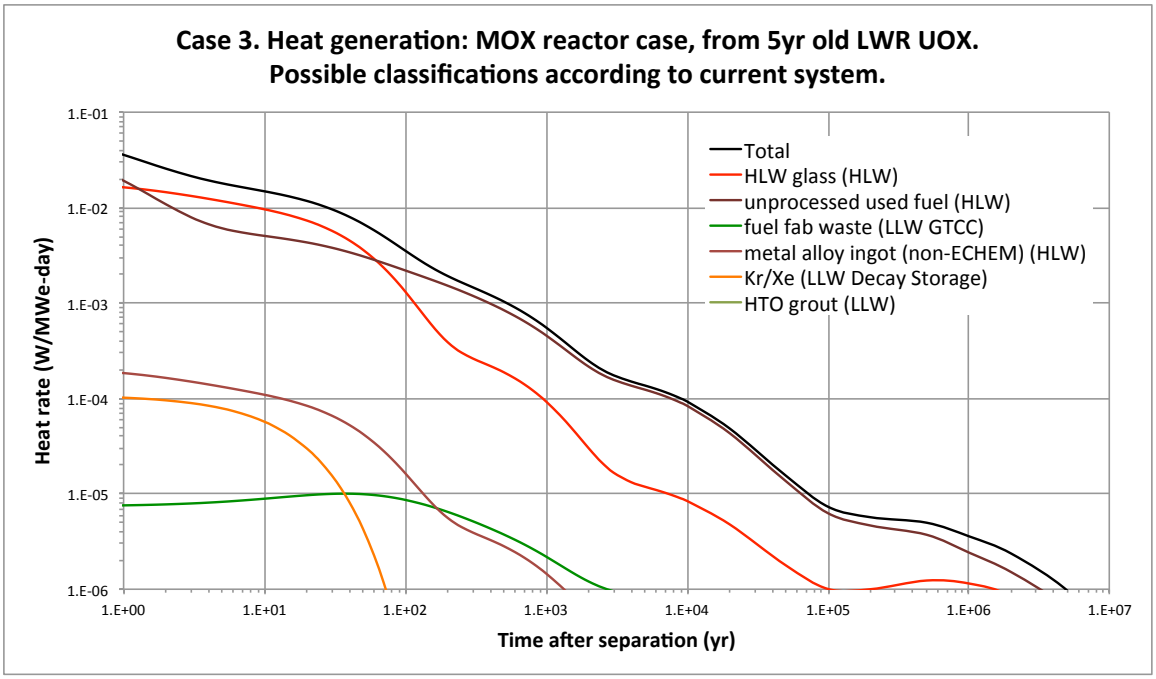

Figure 3.13a: Classification of all waste streams from 5-yr MOX reactor case into existing classes. Normalized to electricity generation per cycle.

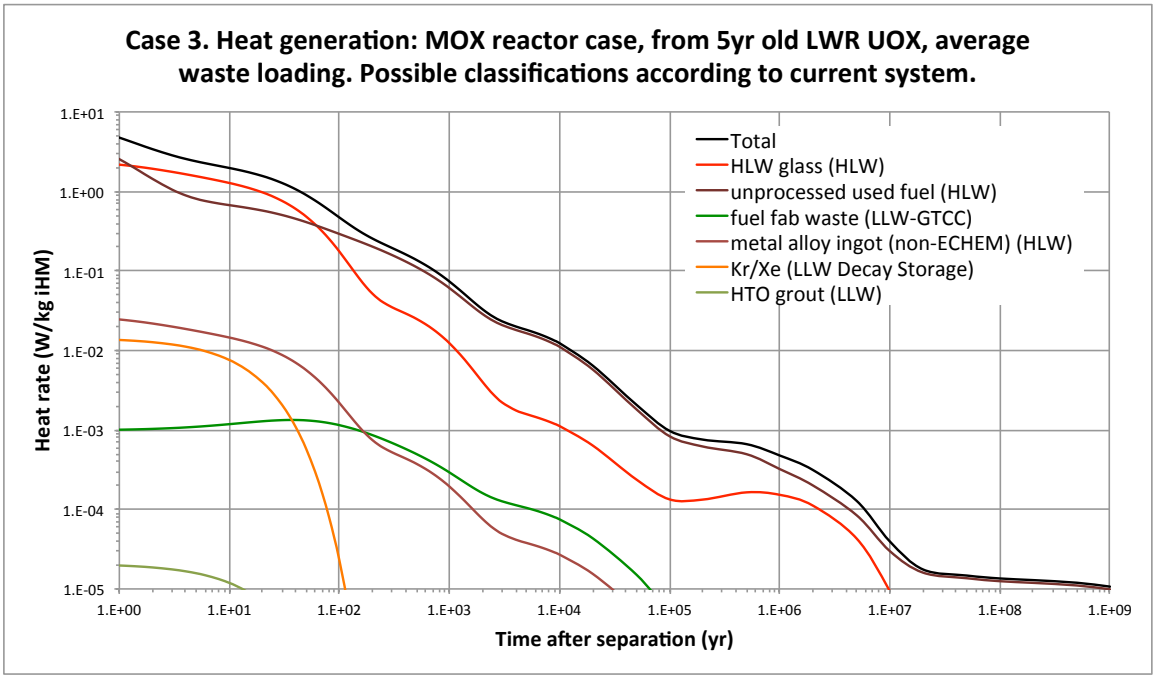

Figure 3.13b: Classification of all waste streams from 5-yr MOX reactor case into existing classes. Normalized to amount of initial heavy metal needed from the initial LWR-UOX reactor per cycle.

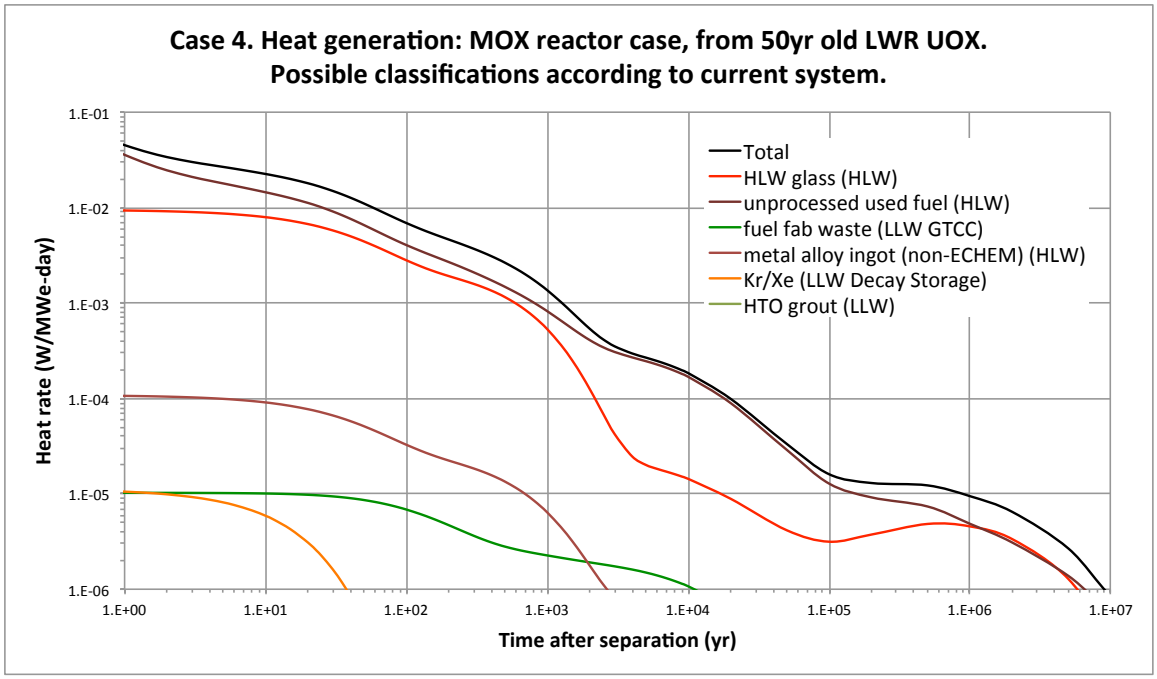

Figure 3.14a: Classification of all waste streams from 5-yr MOX reactor case into existing classes. Normalized to electricity generation per cycle.

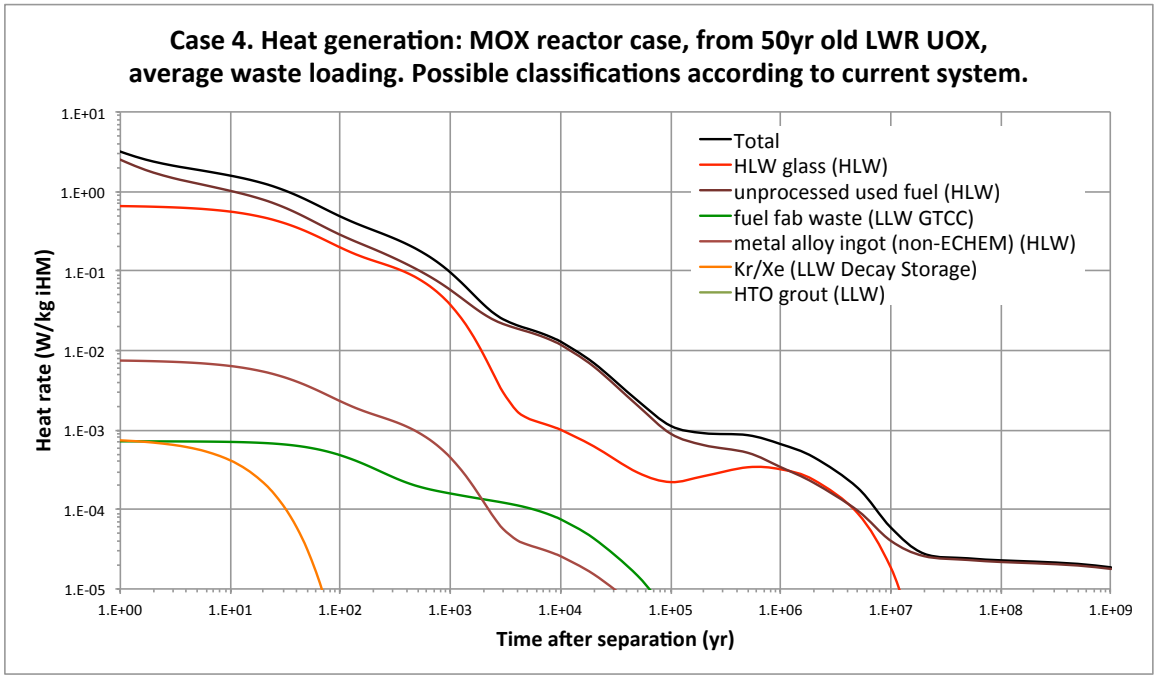

Figure 3.14b: Classification of all waste streams from 5-yr MOX reactor case into existing classes. Normalized to amount of initial heavy metal needed from the initial LWR-UOX reactor per cycle.

For all Figure pairs 3.11—3.14, the relative trends of all curves are similar, independent of the choice of unit normalization.

It is interesting to note that the waste stream with the consistently highest heat content, the unprocessed used fuel bundle, is one of the waste streams with the highest potential for future use, e.g. further reprocessing or recovery of metals for industrial applications. Waste streams such as vitrified HLW glass (from aqueous separations processes), glass-bonded zeolite (fission products from electrochemical reprocessing), and even heat-emitting Cs/Sr, which are all in their final waste form of immobilized separated residual waste, all have orders of magnitude lower heat rate than the final batch of unprocessed used fuel. Low-heat waste such as Kr/Xe could have industrial applications as well. In the case that wastes could be classified according to their potential future use and assigned an corresponding disposal priority, this could be problematic, as spent fuel bundles are difficult to store and cool because of their high heat content.

In conclusion, if all advanced fuel cycle waste streams are classified as HLW, waste disposal benefits of partitioning may be negated. However, even if the current legal framework is not adhered to but the existing classes are kept, an attempt to classify these new waste streams under the current classification system still presents shortcomings. Furthermore, changing current regulations to allow current waste classes to be assigned to more appropriate waste streams, while a step in the right direction, does not take care of classifying all wastes in the entire fuel cycle, such as currently orphaned wastes. Therefore, a catch-all, characteristics-only based classification system is needed.

3.5.2 Classification of FIT Waste under IAEA-Suggested Criteria

The IAEA GSG-1 guidelines classify waste according to activity content and half-life. Because waste streams from the FIT advanced fuel cycle model are expressed in different units, a direct comparison cannot be made, but a qualitative mapping into the IAEA-suggested criteria is possible based on the heat rate data of different waste streams of different fuel cycles. Table 3.6 shows how the IAEA classification guidelines could accommodate every waste stream generated by any of the representative fuel cycles modeled in FIT.

Table 3.6: Possible classification of each waste stream according to IAEA class definitions.

Waste	Suggested class	Meets what criteria from IAEA GSG-1?
Iodine Glass Ceramic	High level waste (HLW)	"Waste with large amounts of long lived radionuclides that need to be considered in the design of a disposal facility for such waste."
Kr-Xe	Very low level waste (VLLW)	"The radionuclides contained in this waste are generally short-lived, although very limited concentrations of longer-lived radionuclides are possible."
Cs-Sr	Low level waste (LLW)	"May include short-lived radionuclides at higher activity levels."
Metal Alloy Ingot (Non-Echem)	Intermediate level waste (ILW)	Contains Tc and other "relatively large concentrations of long-lived radionuclides (generally alpha-emitting radionuclides) that will not decay to a low enough activity level during a reasonable period of time." Requires disposal at greater depths.
Remainder Metal	Intermediate level waste (ILW)	Same as metal alloy ingot.
HTO Grout	Very short lived waste (VSLW)	"This category includes waste that can be cleared from regulatory control by placing it into decay storage for up to a few years."
C14 Grout	Low level waste (LLW)or High level waste (HLW)	Depends on concentration. Fits both definitions of "long-lived radionuclides at relatively low levels of activity concentration" (LLW) and "large amounts of long lived radionuclides that need to be considered in the design of a disposal facility for such waste" (HLW).
LLW Mineralized Monolith	Very low level waste (VLLW)	Same as Kr-Xe.
TRU Mineralized Monolith	Intermediate level waste (ILW)	Same as metal alloy ingot.
Glass	High level waste (HLW)	"Waste with levels of activity concentration high enough to generate significant quantities of heat by the radioactive decay process."
Glass-Bonded Zeolite	High level waste (HLW)	Same as glass.
Fuel Fab Waste	Intermediate level waste (ILW)	Same as metal alloy ingot.
Operational Waste	Very short lived waste (VSLW), Very low level	By definition. Depends on the concentrations and activity levels.

	waste (VLLW), or Low level waste (LLW)	
Operational Waste (TRU)	Intermediate level waste (ILW)	By definition. Contains long-lived TRUs.
Lanthanide Glass	High level waste (HLW)	Ln removed for neutron poison purposes. "Waste with levels of activity concentration high enough to generate significant quantities of heat by the radioactive decay process."
Metal Alloy Ingot (Echem)	Intermediate level waste (ILW)	Contains Tc. Same as metal alloy ingot (Non-Echem).
Unprocessed Used Fuel	High level waste (HLW)	Same as glass.

The levels of contamination of the waste streams from impurities due to separations processes will determine borderline cases. Even in the very low-level waste (VLLW) class, however, there is flexibility within for a presence of long-lived isotopes, so wastes that decay rapidly would not be penalized for possible negligible impurities resulting from processing.

Because the IAEA guidelines are characteristics-based, diverse, and numerous, most waste streams that may result from any separations process can be easily categorized. Furthermore, they can be used to classify wastes orphaned from the current fuel cycle.

3.5.3 "Orphan" Wastes

The current classification framework has left many orphan wastes without a legal framework to deal with its disposition. As a result, a patchwork of current waste classification policies has not adequately dealt with these orphaned wastes, mainly leaving them sitting in aboveground facilities across the United States. Under a classification scheme such as that of IAEA GSG-1, however, these wastes would (and should) be treated differently.

Depleted uranium (DU), as discussed in Chapter 1, did not have a legal framework for disposition or utilization until 2009, when he NRC commissioners voted for DU to be categorized as LLW Class A. However, it had been questioned [NRC 2008] whether it should be classified in a higher LLW category (Class C) due to its longevity, and the decision to classify DU as LLW Class A was controversial [Markey 2009]. Under the IAEA classification framework, it could be either categorized as LLW or IWL, depending on the choice between a long-lived, but very low-level waste. Either way, assigning a class would create a legal imperative to manage it correctly. If possible future utility of the waste was used as informing a classification decision, the scales could tip in favor of classifying DU as a more easily retrievable waste, in the case that DU may be needed for future fast reactor fuel of an advanced fuel cycle fleet.

Mixed low-level waste, or MLLW, currently is a class of waste dealt with only on DOE sites. The IAEA GSG-1 system does not incorporate chemical hazard as thoroughly as the NCRP report #139 does; this could be a useful implementation in the next IAEA waste classification guide.

Greater-than-class-C waste, or GTCC, is currently the de facto "orphan category", although it is not comprehensive by any means. Unfortunately, this leads to some very hazardous wastes being treated for shallow land burial when, based on their characteristics, they should be classified as HLW or ILW waste for deep geologic disposal. A source-based classification does not allow for prudent disposal of even a small fraction of possibly very hazardous wastes.

Regardless of whether the long-term national or global nuclear energy plan involves advanced fuel cycles or not, the existence of orphan wastes alone makes a compelling case for changing the classification system to encompass radioactive wastes of all characteristics and origins. Efforts to address existing isolated low-level waste streams are in progress [NRC 2008]; however, these do not address all wastes and stop short at wastes that could be created in the future. If reclassification of radioactive wastes were already being discussed anyway, a complete revision of the framework would serve long-term public protection much better than a patchwork effort.

Chapter 4

Conclusions and Opportunities for Future Work

Past and ongoing discussions on how to best classify radioactive waste has been a complex task. More recent work, globally and nationally, has supported an evolution from the current source-based classification system to a characteristics- and risk-based one. The legal and political complexities of rewriting the classification framework in the United States are not trivial. However, compelling reasons do exist to do so, not only on the basis of current shortcomings, but also on classification concerns presented by potential future nuclear fuel cycles.

The study presented in this dissertation represents one of the first attempts to examine waste streams from the point of view of advanced fuel cycles with the objective of informing a characteristics-based reclassification effort. It couples wastes generated from a representative set of advanced fuel cycles with previous suggestions of characteristics-based classification systems. The possibility of development and deployment of advanced reactors and the resulting associated processes, leading to the creation of new and different radioactive waste streams, strengthens the need for a classification system that ensures efficient management of those wastes.

Except for within the definitions of LLW and TRU waste, existing waste class boundaries between waste classes are set by the origin of various types of waste. They are not based on specific characteristics such as minimum concentrations, decay heat levels, or external radiation of fission products and long-lived, alpha-emitting transuranic radionuclides. As has been shown, this lets many waste types fall through the cracks. To avoid "orphan wastes", quantitative boundaries are needed, as opposed to qualitative definitions of waste based on source. One of the merits of re-classifying radioactive waste in the United States is the prevention of creation of these orphan wastes. Because these currently have no legal way to be properly managed or disposed, they potentially pose a higher hazard to the environment than higher-activity and -toxicity wastes that are managed properly (an example of this would be mill tailings waste versus spent fuel). It is prudent to link each waste class to a disposal system recommendation to ensure that that each type of waste is properly isolated from humans and the environment. In order to be able do this, however, a classification system that does not leave wastes stranded is needed.

The analysis of waste stream properties from advanced fuel cycles in this dissertation has shown that waste heat is an important metric for informing waste stream classification and disposal choices. The heat metric informs how different high-level waste streams should be treated, as well as provides insights on what waste classes may be needed in a new framework. Waste heat can inform what waste management strategy is needed: decay, disposal, or a combination of both. It is entirely possible that there could be a set of subclasses for different high-level waste streams indicating which short-term strategy is advisable for thermal management of a repository.

This study has also informed, at the very least, the need for the creation of waste classes that accommodate wastes created in advanced fuel cycles that do not share the same characteristics as current high-level waste. A short-lived waste category, such as described in the IAEA classification guidelines, for wastes directly resulting from spent fuel is necessary to accommodate the lower-level separated wastes. Furthermore, wastes created from fuel fabrication processes and to some extent from spent fuel separations can have properties similar to transuranic waste. Rather than being classified as HLW or even GTCC LLW, an intermediate-level waste class that is the equivalent of the defense-only waste TRU class is needed to accommodate the proper disposal of these wastes. If there were a comprehensive characteristics-based category for ILW, DOE and commercial wastes that have similar properties of long half-lives and low radioactivity would not have to be separated. This would be an important advantage especially given the difficulty of licensing a waste disposal site.

Due to the difficulty of waste disposal and repository site licensing, the idea of classifying waste according to its usefulness may be a metric to consider in future work. Being able to rank waste forms according to potential economic value—for example, unprocessed spent nuclear fuel contains untapped energy content as opposed to defense HLW in vitrified glass—could encourage a speedier process towards disposal of wastes that are nothing but exactly that: waste. A "disposal priority" metric coupled with a measure of heat load informing repository management could also be valuable in optimizing between the option of storing certain types of waste in surface facilities or disposing them underground without much further delay. Disposal options such as deep boreholes would be a viable option in the case of high-heat, low-utility waste.

Heat generation of waste produced from fuel cycles normalized to the amount of electricity generated could lead to development of a better measure on how to tax the cost of waste treatment and disposal. The current waste fund fee is arguably an inaccurate representation of the true cost and sends insensitive price signals to waste generators. This could be especially true if advanced reactors with better uranium utilization and lower waste generation (per unit of electricity generated) were part of the future nuclear landscape. Re-evaluating the value of the waste fund fee would in this case be crucial, and classifying waste properly is only ethical based on the fact that the public is being charged with the bill of managing the country's radioactive waste.

Further important future work to strengthen the FIT model includes the ability to evaluate the impact of waste properties of so-called "technology wastes"—wastes incidental to processes in the fuel cycle. This study would have been strengthened this capability would have been incorporated into the model. However, the objective of a catch-all characteristics-based classification system would ensure that whatever the properties of this waste, it would have a framework to be managed properly.

Also, in future modeling work, a repository model incorporating thermal profiles of waste packages and radionuclide migration should be coupled to the fuel cycle model. Usually this is done in order to be able to evaluate the impact of different fuel cycles on repository performance, but in the context of this study, it would also further inform the heat metric analysis performed in this dissertation. Also, exposure pathway models simulating dose rates to humans would better inform classification on metrics such as radiotoxicity and waste form stability, since these indirectly contribute to risk to the public and are strongly dependent on geology.

Considerations of risk must be made on different time scales, and this should be reflected in a characteristics-based classification system. Potential future work on exploring radiotoxicity as a metric in the context of this study, coupled with waste heat studies, could inform which wastes are treated with more weight at what point in time. Generally, waste heat load as a metric should be weighted higher earlier on, whereas wastes that could be problematic from a radiotoxicity viewpoint should be treated importantly later, on much larger timescales. Taking the importance of these waste metrics as a function of time into consideration, a figure of merit could be developed that could further support the development of a new classification framework.

This study has served to inform radioactive waste classification based on insights gained from wastes generated from nuclear fuel cycle processes different from the current fuel cycle. However, regardless of whether advanced fuel cycles are implemented in the near or far future, the need do a better job of protecting people and environment by ensuring prudent radioactive waste management practices is critical.

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