Radioactive waste streams from various potential nuclear fuel cycle options

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Abstract

Five fuel cycle options, about which little is known compared to more commonly known options, have been studied in the past year for the United States Department of Energy. These fuel cycle options, and their features relative to uranium-fuelled light water reactor (LWR)-based fuel cycles, include:

- *Advanced once-through reactor concepts (AOT) – intended for high uranium utilisation and long reactor operating life, use depleted uranium in some cases, and avoid or minimise used fuel reprocessing.*
- *Fission-fusion hybrid (FFH) reactor concepts – potential variations are intended for high uranium or thorium utilisation, produce fissile material for use in power-generating reactors, or transmute transuranic (TRU) and some radioactive fission product (FP) isotopes.*
- *High-temperature gas reactor (HTGR) concepts – intended for high uranium utilisation, high reactor thermal efficiencies; they have unique fuel designs.*
- *Molten salt reactor (MSR) concepts – can breed fissile ^{233}U from Th fuel and avoid or minimise U fuel enrichment, use on-line reprocessing of the used fuel, produce lesser amounts of long-lived, highly radiotoxic TRU elements, and avoid fuel assembly fabrication.*
- *Thorium/^{233}U-fuelled LWR (Th/^{233}U) concepts – can breed fissile ^{233}U from Th fuel and avoid or minimise U fuel enrichment, and produce lesser amounts of long-lived, highly radiotoxic TRU elements.*

These fuel cycle options could result in widely different types and amounts of used or spent fuels, spent reactor core materials, and waste streams from used fuel reprocessing, such as:

- *Highly radioactive, high-burn-up used metal, oxide or inert matrix U and/or Th fuels, clad in Zr, steel, or composite non-metal cladding or coatings.*
- *Spent radioactive-contaminated graphite, SiC, carbon-carbon-composite, metal and Be reactor core materials.*
- *Li-Be-F salt containing U, TRU, Th and fission products.*
- *Ranges of separated or un-separated activation products, fission products and actinides.*

Waste forms now used or studied for used LWR fuels can be used for some of these waste streams – but some waste forms may need to be developed for unique waste streams.

Introduction

A high-level study was performed in Fiscal Year 2009 for the United States Department of Energy (DOE) Office of Nuclear Energy (NE) Advanced Fuel Cycle Initiative (AFCI) to provide information for a range of nuclear fuel cycle options [1]. At that time, some fuel cycle options could not be adequately evaluated since they were not well defined and lacked sufficient information. As a result, five families of these fuel cycle options were studied during Fiscal Year 2010 by the Systems Analysis Campaign for the DOE NE Fuel Cycle Research and Development (FCRD) programme.

The analysis of these fuel cycles also included evaluation of potential waste streams for each option to:

- describe the quality and completeness of the data;
- describe (as practical) waste streams arising from each option;
- identify waste stream similarities and differences (discriminators) for the different options.

The waste stream study relied on the results of the five fuel cycle option studies done separately by the Systems Analysis Campaign. These five potential fuel cycle options are based on the following reactor concepts, which are described in more detail in interim status reports for these studies:

- advanced once-through reactor concepts (once-through) [2];
- fission-fusion hybrid (FFH) reactor concepts [3];
- high-temperature gas reactor (HTGR) [4];
- molten salt reactor (MSR) [5];
- thorium/^{233}U fuelled light water reactor (Th/^{233}U) [6].

Waste streams from the fuel cycle options

Table 1 summarises the radioactive waste streams identified for the different analysed options. Several waste streams (fuel fabrication wastes, reactor core structure materials, and cladding and fuel structure materials) would be radioactive due to contamination with radioactive fuel components, fission products or activation products. This radioactivity will affect their recyclability and impact disposal requirements. Front-end radioactive wastes from mining and enrichment are listed for completeness. Amounts and concentrations of radioactive materials in these waste streams are expected to be much less than in used fuel, but the amounts of these wastes will vary for some of the fuel cycle options – for example, some options do not require U fuel enrichment, so depleted uranium would be eliminated in those options.

Noble gases (mainly xenon and krypton-85), iodine-137, carbon-14 and tritium are grouped together in the table as “gaseous fission products (FP).” Gaseous FP would evolve from all reprocessing options, and would be separately captured and immobilised in separate waste forms designed for the chemistry and to meet the disposal requirements for each of these gases, even though they are grouped together in the table.

Semi-volatile and non-volatile FP can be separately captured and immobilised in separate waste forms, if desired, in some fuel cycle options. In other fuel cycle options, these FP are only separated from recycled fuel in a single waste stream and immobilised in a single waste form.

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Table 1: Radioactive waste streams expected from some potential fuel cycle options

Fuel cycle option and family		Fuel cycle category	Candidate separation process	Radioactive waste streams [yes (Y) = expected, no (N) = not expected] for that fuel cycle option										
				U/Th mining	U enrich-ment	Fuel fabrication	Core structure	Cladding and fuel structure	Gaseous FP	Semi-volatile FP	Non-volatile FP	Acti-nides	Spent fuel assemblies, particles, pins	Comments
Advanced once-through reactor concepts														
1	Ultra Long Life Fast Reactor (ULFR)	Mod. open	Aqueous, echem	Y	Y	Y	N	N	N	N		N	Y	
2	CANDLE	Once-through	–	Y	Y	Y	N	N	N	N	N	N	Y	
3	Fast Mixed Spectrum Reactor (FMSR)	Once-through	–	Y	Y	Y	N	N	N	N	N	N	Y	Same as CANDLE.
4a	Travelling Wave Reactor (TWR)	Once-through	–	Y	Y	Y	N	N	N	N	N	N	Y	
4b	Travelling Wave Reactor (TWR)	Mod. open	Melt refining	Y	Y	Y	N	Y	Y	Y	Y		Y	Melt refining, being a less efficient separations process, will result in actinide contamination in FP waste streams.
5a	Energy Multiplier Module (EM²)	Once-through	–	Y	Y	Y	N	N	N	N	N	N	Y	
5b	Energy Multiplier Module (EM²)	Mod. open	DUPIC	Y	Y	Y	N	Y	Y	N	N	N	Y	DUPIC chops but does not heat or dissolve SNF. In this DUPIC variant, cladding and gaseous FP are removed and disposed.

Echem = Electrochemical.

LWR = Light water reactor.

CANDLE = Constant axial shape of neutron flux, nuclide density and power shape during life of energy production.

Mod. open = Modified open cycle.

DUPIC = Direct use of spent PWR fuel in CANDU.

SNF = Spent nuclear fuel.

CANDU = Canada deuterium uranium reactor.

Table 1: Radioactive waste streams expected from some potential fuel cycle options (*cont.*)

	Fuel cycle option and family	Fuel cycle category	Candidate separation process	Radioactive waste streams [yes (Y) = expected, no (N) = not expected] for that fuel cycle option										Comments
				U/Th minin g	U enrich-ment	Fuel fabrication	Core structure	Cladding and fuel structure	Gaseous FP	Semi-volatile FP	Non-volatile FP	Acti-nides	Spent fuel assemblies, particles, pins	
Fusion-fission Hybrid (FFH) reactor														
1	CFNS coupled with FFTS waste burner	Full recycle	Aqueous, echem	N	N	Y	Y	Y	Y	Y		N	N	Burns waste from LWR used fuel reprocessing, multi-recycle. Assumes the U mining/enrichment is attributed to the LWR.
2a	Fission Suppressed Breeder (FSB) fissile fuel factory	Full recycle	On-line	N	N	N	Y	Y	Y	Y		N	N	Assumes DU fuel, and wastes from U mining/enrichment is attributed to other reactors that need EU. Many variations are possible depending on U or Th fuel, and how bred U/TRU is cycled in other reactors.
2b	Fission Suppressed Breeder (FSB) fissile fuel factory	Full recycle	On-line	Y	N	N	Y	Y	Y	Y		N	N	Assume Th-fuelled, so wastes from Th mining are included in this analysis.
3a	Laser Inertial Fusion Energy (LIFE) once-through deep burn concept	Once-through	–	Y	N	N	Y	N	N	N		N	Y	This once-through variant assumes that gaseous FP are retained in the fuel.
3b	Laser Inertial Fusion Energy (LIFE) once-through deep burn concept	Mod. open	On-line	Y	N	N	Y	Y	Y	N		N	Y	This mod. open variant assumes that gaseous FP are released from the fuel, captured, and disposed; and some cladding/fuel structure is discarded.

CFNS = Compact fusion neutron source fusion driver.

FFTS = Fission fusion transmutation system.

DU = depleted uranium.

EU = enriched uranium.

TRU = transuranic.

Table 1: Radioactive waste streams expected from some potential fuel cycle options (*cont.*)

	Fuel cycle option and family	Fuel cycle category	Candidate separation process	Radioactive waste streams [yes (Y) = expected, no (N) = not expected] for that fuel cycle option										
				U/Th mining	U enrich-ment	Fuel fabrication	Core structure	Cladding and fuel structure	Gaseous FP	Semi-volatile FP	Non-volatile FP	Acti-nides	Spent fuel assemblies, particles, pins	Comments
High-temperature Gas Reactor (HTGR)														
1a	Once-through HTGR	Once-through	–	Y	Y	Y	Y	N	N	N		N	Y	A TRISO fuel particle that retains gaseous FP is assumed.
1b	Once-through HTGR	Once-through	–	Y	Y	Y	Y	N	Y	N		N	Y	BISO fuel is assumed that vents gaseous FP.
2	Minimal fuel treatment HTGR	Mod. open	AIROX	N	N	Y	Y	Y	Y	Y			Y	Recycles LWR used fuel; limited separations; attribute U mining/enrichment to LWR.
3	Single recycle in HTGR	Mod. open	Aqueous or echem	N	N	Y	Y	Y	Y	Y		N	Y	First reactor could be LWR or HTGR; full separations.
4	Sustained recycle with HTGR only	Full recycle	Aqueous or echem	N	N	Y	Y	Y	Y	Y		N	N	Full separations.
Molten Salt Reactor (MSR)														
1	Two-fluid Th MSR	Full recycle	On-line	Y	N	N	Y	N	Y	Y		N	N	Full separations.
2	Single-fluid Molten Salt Breeder Reactor (MSBR)	Mod. open or Full recycle	On-line	Y	N	N	Y	N	Y	Y		N	N	Full separations.
3	Single-fluid Denatured Molten Salt Breeder Reactor (DMSBR)	Once-through	–	Y	Y	N	Y	N	Y	Y	N	N	Y	^{238}U is used for denaturing; sparging to remove gaseous and noble FP.
Thorium/^{233}U-fuelled Light Water Reactors (Th/^{233}U)														
1	Th/^{233}U fuel multi-recycle in current PWR	Full recycle	Aqueous or echem	Y	N	Y	N	Y	Y	Y		N	N	No U used after initial start-up.

AIROX = Atomics International Reduction Oxidation.

Full recycle systems maximize actinide utilisation. Once-through and modified open systems will have disposed actinides in spent fuel or in separated waste streams from reprocessing.

All fuel cycles may have about the same amounts of FP per unit of energy produced. Designs to transmute some FP exist; the FFH waste burner is the only one included in this analysis.

Most of the fuel cycle options are insufficiently defined to specify types and amounts of wastes from fuel fabrication, reactor cores and cladding/fuel structure.

Wastes produced to provide starting fissile material, and reactor decommissioning wastes after closure, exist but are not included here. Fabrication facility, reactor operation and separations facility operations and maintenance LLW can be significant but are not included here. Reactor core wastes include metallic and non-metallic core structures, moderators and reflectors that must be periodically discarded and replaced. Fuel structure and cladding includes fuel assemblies, support structures, inert components of IMF, coatings on particles and pebbles, and molten salts.

Advanced once-through reactor concepts

Advanced once-through fuel cycle concepts are designed to achieve higher U utilisation than is typical for UOX-fuelled LWR, and avoid reprocessing used fuel. Several AOT variations exist; five have been included in the AOT study [2]. All of these variations use enriched U fuel, and so will generate front-end radioactive wastes from U mining and enrichment. All five use fast reactors and use either fuel shuffling or separate zones that contain fissile and fertile isotopes, so that larger amounts of both the fissile and fertile isotopes are eventually burned. The fertile material is either natural or depleted U; the starting fissile material is either transuranic (TRU) material separated and recycled from LWR used fuel, or EU; and fissile material for continued operation after a start-up time period is either bred in the AOT reactor, or else obtained from recycled used LWR fuel.

Once-through fuel cycle concepts do not produce waste streams from separating used fuel (because no reprocessing is done), and only produce spent fuel that is direct-disposed. These spent fuels will contain gaseous and other fission products and un-burned actinides. They will have high short-term radiolytic heat generation and high initial radiotoxicity because of high levels of FP in the spent fuel, because higher fuel burn-up results in higher levels of FP. But long-term heat generation and radiotoxicity should be relatively lower, because the expected levels of long-lived TRU should be relatively lower.

Two variations included in the AOT analyses illustrate how such concepts as the Travelling Wave Reactor (TWR) and the Energy Multiplier Module (EM^2) can also include limited recycling designed to further increase uranium utilisation and better utilise residual TRU, while still minimising potential proliferation concerns that can occur for full recycling options. In these variations, melt refining or other limited recycling technologies may be able to remove enough FP from used fuel to enable the remaining fuel material to be recycled to a reactor; or the used fuel may be chopped and packaged into a fuel bundle for a Canada Deuterium Uranium (CANDU) reactor, without any chemical reprocessing, in a DUPIC (Direct Use of Spent PWR fuel in CANDU) recycle process. Both of these variants will cause some evolution of gaseous FP (in DUPIC) and evolution of a wider range of gaseous, semi-volatile and relatively non-volatile FP (in melt refining), also likely contaminated with relatively smaller amounts of actinides. These variations will result in the evolution of some waste streams from the reprocessing operations, and cause these fuel cycle concepts to be more appropriately described as “modified open” rather than “once-through” options.

Fusion-fission hybrid

Waste streams from FFH concepts are not well qualified or quantified because no FFH systems currently exist and their technical maturity is in early stages. However, the FFH options in this study can result in radioactive waste scenarios that are unique compared to other fuel cycle options. Specific waste streams that may be more unique to FFH options compared to other fuel cycle options include:

- tritium contamination and/or losses;
- heavily irradiated beryllium metal;
- salt processing wastes including tritium and beryllium contamination;
- structural components activated by high energy neutrons.

Three FFH options have been addressed in the FFH study [3]. The dedicated FFH waste burner is designed to reduce the waste disposal challenges from the larger LWR energy production fleet, primarily through elimination of most actinides from the waste. Its actinide burning efficiency is expected to be high enough that it is likely that amounts of MA that are eventually discarded will be mainly limited to used fuel processing losses in both the LWR recycle and the FFH recycle. It is also possible to transmute significant portions of long-lived fission products if deemed desirable – at a cost in neutron economy in the FFH burner.

The Fission Suppressed Breeder (FSB) fissile fuel factory seeks to produce minimal high-level radioactive waste (HLW) via suppression of fission in the breeding blanket. Fission products may be removed in on-line salt processing or left to accumulate in the blanket. The largest amounts of radioactive wastes may result from how the fissile material produced in the FFH fissile factory is used, in once-through, modified open or full recycle options. Potential waste streams from two different FSB options [based on whether depleted uranium (DU) or thorium is used as the fuel] are included in this analysis.

The Laser Inertial Fusion Energy (LIFE) once-through deep burn option seeks to minimise TRU sent to waste by pushing the burn-down phase as far as needed to meet waste management objectives. Waste quantities are minimised through maximum energy extraction from the fissionable resource. With higher resource utilisation, the fission product levels will also be higher (in rough proportion with the resource utilisation and fuel burn-up). The spent fuel would have fission product concentrations much higher than is typical in LWR used fuel, which must be considered in packaging, storage, and disposal facility heat management.

High-temperature gas reactor

High temperature gas reactor options reviewed in this study illustrate a range of potential once-through, modified open and full recycle categories and include those that would use a range of Tristructural-isotropic (TRISO) and Bistructural-isotopic (BISO) U, TRU and mixedoxide (MOX) fuels. The different fuel cycle categories and different potential fuels imply different potential radioactive waste streams and compositions. Radioactive waste streams in HTGR concepts that are not common in most other fuel cycle options include:

- graphite blocks from reactor cores;
- SiC and C coatings separated from fuel particles and pebbles during recycle;
- discarded spent fuel in once-through cases (particles or pebbles coated with durable SiC and C (or other material) coatings.

A few waste management issues are unique to HTGR options. The graphite block moderator material could be a relatively large-mass radioactive waste stream compared to other HTGR radioactive waste streams, unless the graphite material can be recycled. Analyses and planning have been done to determine how to best recycle this material. In addition, the coatings on fuel particles and pebbles, designed for durability and toughness, present a challenge during reprocessing. These coatings also represent a large-mass waste stream in fuel recycling options.

Molten salt reactor

MSR variations can include different fuels (enriched uranium [EU] or Th), single or two-fluid molten salt designs, or operation with or without on-line reprocessing. These variations result in variations in potential types and amounts of waste streams which are not common to most other fuel cycle options, including:

- no cladding;
- graphite blocks from reactor cores;
- spent molten salt fuel in the once-through denatured molten salt breeder reactor (DMSBR) case.

The continuous on-line separations in all MSR recycle options and the continuous gaseous and noble metal sparging in the once-through option enable higher burn-up than in other non-MSR options because of the removal of FP poisons and the lack of cladding that can degrade over time under high burn-up conditions. So, like other high burn-up options, levels of FP produced during MSR operation and collected in waste streams separated during online reprocessing will be relatively high, in proportion with the fuel burn-up; but unlike other cases, levels of fissile materials and TRU in the fuel need not be high, since levels of FP poisons can be continuously maintained at lower levels than would occur in other high-burn-up fuels.

Waste streams containing semi-volatile and non-volatile FP separated during on-line reprocessing can be quite concentrated, and can contain amounts of salt waste, and so will require consideration of heat generation and waste loadings during subsequent handling and management.

Thorium/^{233}U multi-recycle in pressurised water reactor

The Th-fuelled pressurised water reactor (PWR) multi-recycle fuel cycle avoids uranium enrichment because it uses Th fuel in normal operation. The long-term heat and radiotoxicity of waste streams from the ^{233}Th/^{233}U fuel cycle are less compared to the ^{238}U/^{239}Pu fuel cycle, because smaller amounts of TRU elements are produced. Still, other isotopes including ^{231}Pa, ^{229}Th and ^{230}U are produced which must be included in waste radiotoxicity and proliferation risk analyses.

Irradiated THO_2-based fuels contain ^{232}U, which has strong gamma-emitting daughters ^{212}Bi and ^{208}Tl, and which aids in proliferation mitigation but causes remote, shielded and automated reprocessing and re-fabrication. However, proliferation risk considerations should not be minimised. ^{233}U is fissile and could be misused. One possible proliferation-mitigating solution, denaturing the fuel with ^{238}U, increases the normally undesirable production of TRU elements. In addition, proliferation risk will continue to exist from separations processes needed for multi-recycle that could produce recycle streams that are candidates for misuse.

Results and conclusions

Results and conclusions include:

- Families of several fuel cycle options cross-cut across the once-through, modified open and full recycle strategies.
- Limited fuel reprocessing such as DUPIC, AIROX or melt refining will generate some radioactive wastes including fuel cladding and structure materials, gaseous fission products, and (in some cases) semi-volatile fission products. Limited fuel reprocessing will likely result in less efficient separations of waste FP from recyclable actinides, resulting in waste FP contamination in recycled actinide streams and TRU contamination in waste FP streams.
- Fission product contamination of recycled fuel, and the presence of TRU elements, will cause recycled fuel handling and fabrication operations to be remote operations inside shielded hot cells.
- TRU contamination in some waste streams will cause those streams, which otherwise might meet LLW Class C limits, to require disposition as GTCC LLW.
- The amounts of radioactive non-fuel wastes from reactor core structures and fuel cladding and structure materials for some fuel cycle options can be large compared to the used fuel waste streams. Some analyses have been done to evaluate how to recycle these relatively large waste streams.
- High-burn-up used fuels will have high concentrations of high-heat-generating and high-radiotoxicity isotopes, that may cause lower waste loadings in waste forms and in geological repositories to stay within expected thermal and radiotoxicity limits.
- Full recycle options can significantly lower short- and long-term radiotoxicity and heat generation compared to some once-through and modified open options, because of the transmutation of high-radiotoxicity, high-heat, long-lived TRU isotopes.

The quality and completeness of data available to date for the fuel cycle options is insufficient to perform quantitative radioactive waste analyses using recommended metrics. This study has been limited thus far to qualitative analyses of waste streams from the candidate fuel cycle options, because quantitative data for wastes from the front end, fuel fabrication, reactor core structure, and used fuel for these options is generally not yet available. These data gaps exist for most of the fuel cycle options evaluated in this study. At the time such data are available, these additional waste stream analyses can be done:

- mass, volume and compositions of different radioactive waste streams;
- mass, volume and waste loading of waste forms for the different waste streams;
- radiotoxicity and heat generation of the radioactive wastes.

The mass, volume, composition, radiotoxicity and heat generation for the waste streams and waste forms can be normalised to the amount of thermal or electric energy produced for the different options.

References

[1] Wigeland, R., *et al.*, *AFCI Options Study*, AFCI-TIO-PMO-MI-DV-2009-000086, Advanced Fuel Cycle Initiative, September (2009).

[2] Taiwo, T.A., T.K Kim, *Interim Report on Once-through Fuel Cycle Systems*, Argonne National Laboratory, ANL-FCRD-303, 31 March (2010).

[3] Halsey, W., *Interim Status Report – Fusion-fission Hybrid Technology Assessment*, Lawrence Livermore National Laboratory, 31 March (2010).

[4] Piet, Steven J., *et al.*, *Status Report – HTGR Technology Family Assessment for a Range of Fuel Cycle Missions*, Idaho National Laboratory, 31 March (2010).

[5] Gehin, Jess, Lou Qualls, Kent Williams, *Interim Report on Molten Salt Reactor Fuel Cycle Options*, Oak Ridge National Laboratory, 31 March (2010).

[6] Taiwo, T.A., D. Yun, T.K Kim, *Interim Report on Th/^{233}U Multirecycle in PWR*, Argonne National Laboratory, ANL-FCRD-304, 31 March (2010).