Improving the Estimates of Waste from the Recycling of Used Nuclear Fuel - 13410

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ABSTRACT

Estimates are presented of wastes arising from the reprocessing of 50 GWD/tonne, 5 year and 50 year cooled used nuclear fuel (UNF) from Light Water Reactors (LWRs), using the "NUEX" solvent extraction process. NUEX is a fourth generation aqueous based reprocessing system, comprising shearing and dissolution in nitric acid of the UNF, separation of uranium and mixed uranium-plutonium using solvent extraction in a development of the PUREX process using tri-n-butyl phosphate in a kerosene diluent, purification of the plutonium and uranium-plutonium products, and conversion of them to uranium trioxide and mixed uranium-plutonium dioxides respectively. These products are suitable for use as new LWR uranium oxide and mixed oxide fuel, respectively. Each unit process is described and the wastes that it produces are identified and quantified. Quantification of the process wastes was achieved by use of a detailed process model developed using the Aspen Custom Modeler suite of software and based on both first principles equilibrium and rate data, plus practical experience and data from the industrial scale Thermal Oxide Reprocessing Plant (THORP) at the Sellafield nuclear site in the United Kingdom. By feeding this model with the known concentrations of all species in the incoming UNF, the species and their concentrations in all product and waste streams were produced as the output. By using these data, along with a defined set of assumptions, including regulatory requirements, it was possible to calculate the waste forms, their radioactivities, volumes and quantities. Quantification of secondary wastes, such as plant maintenance, housekeeping and clean-up wastes, was achieved by reviewing actual operating experience from THORP during its hot operation from 1994 to the present time. This work was carried out under a contract from the United States Department of Energy (DOE) and, so as to enable DOE to make valid comparisons with other similar work, a number of assumptions were agreed. These include an assumed reprocessing capacity of 800 tonnes per year, the requirement to remove as waste forms the volatile fission products carbon-14, iodine-129, krypton-85, tritium and ruthenium-106, the restriction of discharge of any water from the facility unless it meets US Environmental Protection Agency drinking water standards, no intentional blending of wastes to lower their classification, and the requirement for the recovered uranium to be sufficiently free from fission products and neutron-absorbing species to allow it to be re-enriched and recycled as nuclear fuel. The results from this work showed that over 99.9% of the radioactivity in the UNF can be concentrated via reprocessing into a fission-product-containing vitrified product, bottles of compressed krypton storage and a cement grout containing the tritium, that together have a volume of only about one eighth the volume of the original UNF. The other waste forms have larger volumes than the original UNF but contain only the remaining 0.1% of the radioactivity.

INTRODUCTION

As part of their past work funded by the US Department of Energy (DOE) for the Global Nuclear Energy Partnership (GNEP), several industry groups, including one led by Energy*Solutions*, developed conceptual designs and process flowsheets for a US facility to recycle used nuclear fuel (UNF) from the US fleet of Light Water Reactors (LWRs). The aqueous-based facility designed by the Energy*Solutions* team was capable of processing UNF at a rate of 5 tonnes per day, at burn-ups up to 50 GWD per tonne of UNF and a minimum of 5 years out-of-reactor cooling. The facility was designed to produce (i) a pure uranium stream which was to be re-enriched and re-used in LWRs, or used directly in Heavy Water moderated reactors, (ii) a mixed uranium-plutonium (and optionally neptunium) stream which was to be converted directly into Mixed Oxide (MOX) fuel and re-used in LWRs, (iii) an optional separated minor actinide (americium and curium) stream that was to be formed into targets and burned in either thermal or

fast neutron reactors so as to keep these long lived heat-emitting species out of the waste repository, and (iv) a high level waste stream which would be vitrified ready for permanent disposal. The facility was based on third generation European recycling technology (on a scale where the pioneering US Hanford and Savannah River facilities are first generation) and, with further improvements enabled by continually improving technology, would be fourth generation. It was capable of being built and put into use on a short timeframe (<20 years) in the US and was also part of an overall plan that would bring fast neutron reactors into the scheme over a longer 50+ year timeframe.

As part of this design work, the Energy*Solutions* team estimated all primary and secondary wastes that such a recycling facility would produce. These comprise (i) the vitrified high level waste, captured krypton-85 and tritium (containing over 99.9% of all the radioactivity in the UNF), (ii) Class C and Remote Handled Greater Than Class C wastes (containing almost all of the rest of the radioactivity, and formed of the UNF zircaloy cladding plus salt wastes and certain captured volatile fission products), and (iii) secondary wastes from, for example, facility maintenance and cleanup work.

Differences between these waste estimates, those made by the other industry teams, and ones made by some of the US National Laboratories were subsequently identified. It was recognized at that time that the significant differences could be traced to a range of differing assumptions made by each team in their bases of design for the recycling facility and in the assumed regulatory climate.

To address and better understand differences in waste estimate generation when analyzing various potential future fuel cycle options, DOE has recently placed task orders under existing contracts with Energy*Solutions* and with another industry team to conduct a study to determine more precisely waste arising from a UNF recycle facility using a common set of high-level assumptions, and with review support provided by US National Laboratories. The work that the Energy*Solutions* team initially carried out under this task order is described, and a summary of the waste estimates produced is provided. These initial waste estimates may be revised as the task work proceeds.

WASTE ESTIMATING SCENARIOS

The task order required the estimation of wastes arising from three UNF reprocessing and three nuclear fuel fabrication scenarios. Reported here are the waste estimates arising from two of these scenarios:

(i) Reprocessing of LWR uranium oxide fuel at a burnup of 50 GWD/tonne and an out-of-reactor cooling time of 5 years.
(ii) Reprocessing of LWR uranium oxide fuel at a burnup of 50 GWD/tonne and an out-of-reactor cooling time of 50 years.

Scenario (ii) recognizes that, given the reality of extended UNF storage times prior to reprocessing, a new reprocessing facility might reprocess existing long-cooled fuel for a substantial period of time, and thus benefit from the decay of several shorter half-life fission products that otherwise complicate waste treatment. However, scenario (i) recognizes that, should a reprocessing facility be constructed, it will likely be desirable for it to also have a capability to process shorter-cooled fuel. This fuel is likely to be stored in water-filled pools, and reprocessing it first leaves in place the longer-cooled fuel that is already safely contained in the dry storage casks that are now in routine use by the power utilities.

BASIS OF THE REPROCESSING WASTE ESTIMATE

NUEX and THORP Reprocessing

The basis of the waste estimates produced by this work was the "NUEX" reprocessing chemical flowsheet (Figure 1) produced by Energy*Solutions* for the DOE sponsored Global Nuclear Energy Partnership (GNEP) project. This flowsheet and the process equipment to operate it [1, 2] is a fourth generation development of the "PUREX" solvent extraction reprocessing technology using tri-n-butyl phosphate (TBP), and pioneered in the 1940s and 1950s by the Hanford and Savannah River sites in the

U.S.A. NUEX is itself built upon the third generation reprocessing technology developed for the Thermal Oxide Reprocessing Plant (THORP) at Sellafield in the United Kingdom [3]. THORP is an industrial scale reprocessing plant capable of processing 5 tonnes of UNF per day and producing three main outputs.
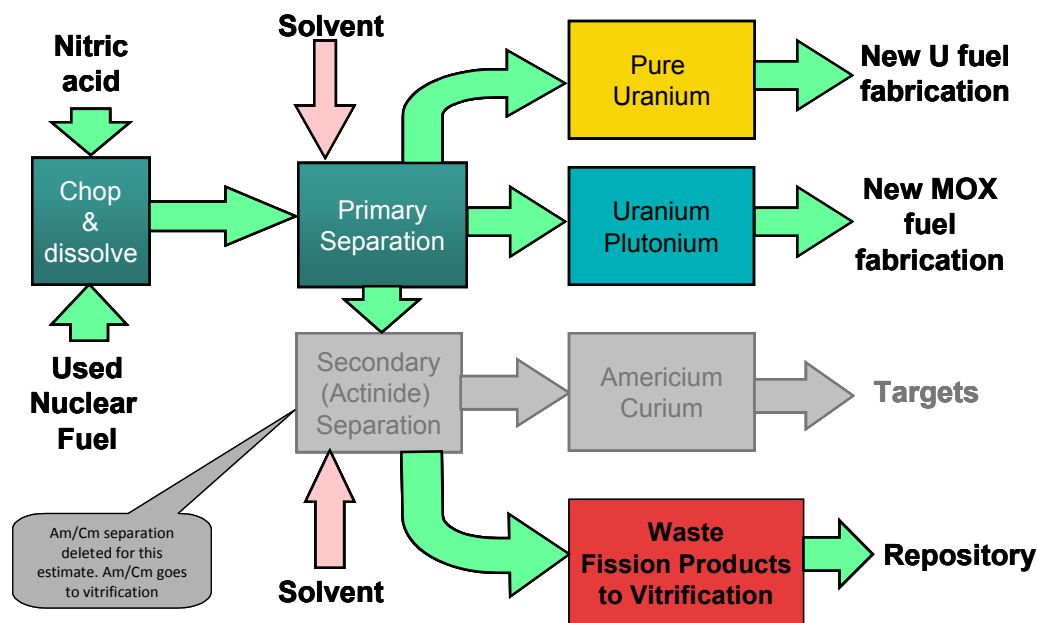
Figure 1 Outline of the NUEX Process

These are purified uranium as uranium trioxide, purified plutonium as plutonium dioxide, and fission product wastes which are vitrified and contain over 99% of the radioactivity originally present in the UNF. THORP started hot operations in 1994 and is currently in full commercial operation.

The process equipment planned during the GNEP period for the NUEX reprocessing plant is nearly identical to the well-proven equipment in current use in THORP. This was a deliberate decision to minimize the need for fresh equipment development work. The NUEX process flowsheet differs from THORP, however, in three major respects (Figure 1):

- Pure plutonium is not separated at any point in the process but instead stays mixed throughout the process with some uranium. This is a fissile material security measure [4, 5], the uranium mixed with the plutonium making it less attractive for potential weapons production.
- The gaseous fission products iodine-129, carbon-14, krypton-85 and hydrogen-3 (tritium) are all captured and consigned as waste. For the GNEP study this was in recognition of the anticipated regulatory environment for any new reprocessing facility in the U.S.A. For the present study it served to allow assessment of the waste impact associated with this capture. In THORP only the I-129 and C-14 are captured, with the Kr-85 allowed to disperse into the atmosphere from a tall stack and the tritium discharged to the ocean, both allowed under UK regulatory limits that were in place when THORP was designed and commissioned.
- The minor actinides americium and curium are captured from the fission product waste stream in NUEX by additional cycles of solvent extraction and are consigned as targets for future destruction in fast reactors – and possibly also in thermal reactors [6,7,8]. This modification was introduced for GNEP to keep these very long-lived heat emitters out of the vitrified product, allowing a denser packing of the glass 'logs' in a future high level waste repository. However, this provision was not continued in the flowsheet used for the current waste estimating exercise.

These differences are shown in more detail in Figure 2, with the americium, curium removal steps grayed out because they were not part of the current waste estimate work.
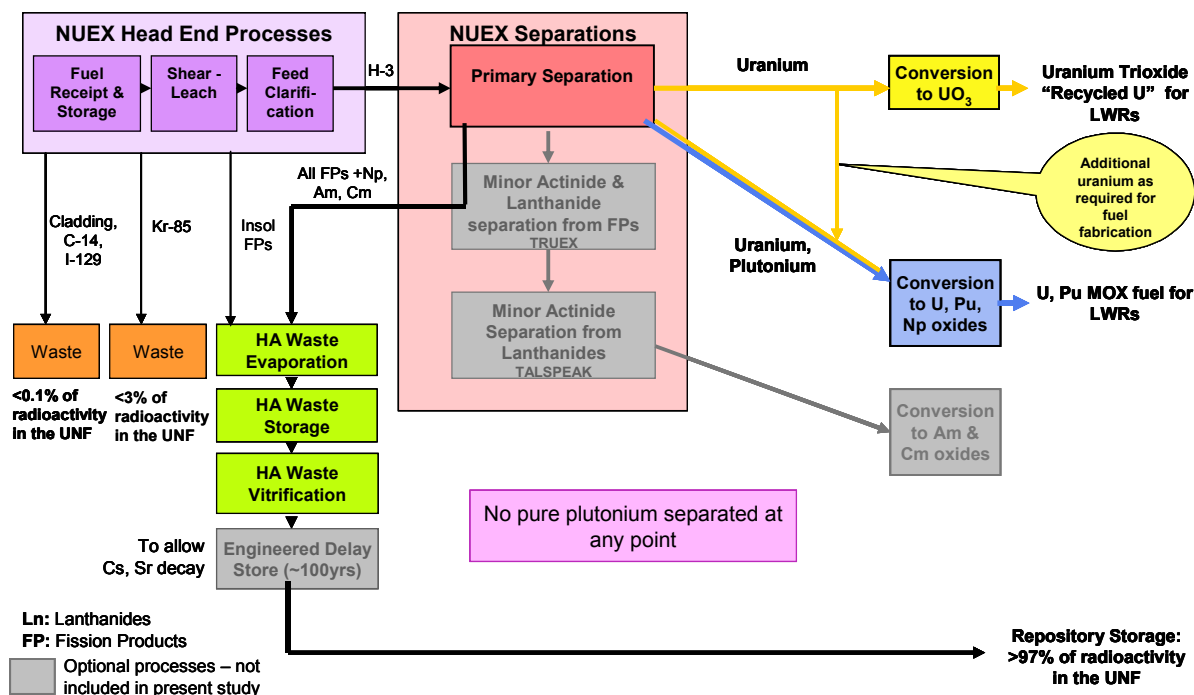

Figure 2 More Details of the NUEX Reprocessing Flowsheet

DESCRIPTION OF THE NUEX REPROCESSING PROCESSES AND THEIR WASTES

Fuel Receipt and Storage

Used nuclear fuel enters the facility at Fuel Receipt and Storage where it is stored in Multi-Purpose Canisters (MPCs), themselves contained within shielded dry casks. When selected for reprocessing, the fuel assemblies are removed from their containment under water in a feed basin and then transported within a shielded enclosure ("cave") out of the water into the shear cave. Wastes generated from these steps include used MPCs each assumed to initially contain 10 tonnes of fuel, fuel pool water treatment materials such as ion exchange media and filters, plus ad-hoc maintenance wastes. A summary of wastes from all parts of the NUEX process is shown in Figure 3.

Head End Fuel Shearing

The UNF assemblies are sheared into 5 cm (2 inch) sections by moving them progressively under a vertically operating shear blade. This action cuts the individual fuel pin zircaloy cladding, thus exposing the oxide fuel pellets within. The chopped sections of fuel pin fall under gravity into a perforated basket in one of several fuel dissolvers. The main wastes from fuel shearing are used shear blades, which must be replaced fairly frequently to ensure clean cutting of the fuel pins without crimping, which would inhibit efficient dissolution, plus ad-hoc and maintenance materials wastes.

Head End Fuel Dissolution and Feed Clarification

The sheared fuel pin sections are contacted with nitric acid in the dissolvers, which dissolve (or leach) the oxide fuel material away from the zircaloy cladding "hulls". This results in four output streams: (i) the dissolved UNF, (ii) undissolved fission products and fuel cladding particles, (iii) the cladding hulls, and (iv) off-gases from the dissolution process. The dissolved UNF and undissolved fission products are
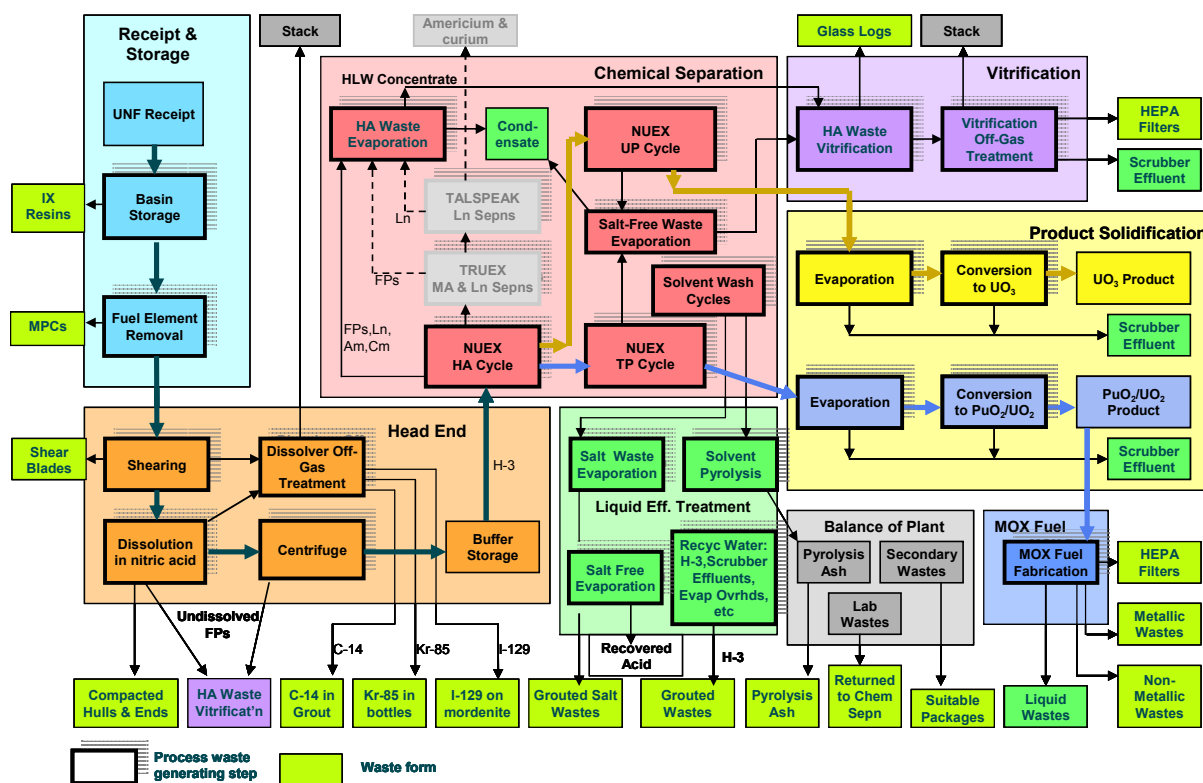
Figure 3 – Wastes Generated by the NUEX Process

separated from each other in one of several centrifuges, with the clarified dissolver solution being fed forward, via buffer storage, to Chemical Separation, and the slurry of undissolved fission products being combined with the high level waste from Chemical Separation and sent for vitrification. The cladding hulls are removed in the dissolver basket and, after washing with nitric acid and compaction, are consigned as a Remote Handled Greater Than Class C (RH-GTCC) waste stream. The dissolver off-gases contain all the Kr-85, and most (typically ~98%) of the I-129 and C-14 that was in the UNF. The Kr-85 (and non-radioactive krypton isotopes) are removed by cryogenic distillation and stored under pressure in cylinders until the Kr-85 radioactive decay (half-life 10.7yrs) allows all the krypton to be discharged. The I-129 is sorbed onto silver mordenite solid and consigned as GTCC waste. An encapsulant for the I-129 on silver mordenite has not yet been defined and is the subject of ongoing research and development. The C-14 is removed by sodium hydroxide scrubbing and then reacted with calcium nitrate to precipitate calcium carbonate which is grouted in cement. This waste is classified Class C, not because of its C-14 content but because of traces of UNF dust that contaminate it. For this reason, an option is to combine the grouted C-14 waste with the UNF cladding hulls and I-129 as a RH-GTCC waste. Tritium is also evolved as tritiated water vapor into the dissolver off-gas during dissolution but is returned to the dissolver solution by the off-gas scrubbers and so moves with the dissolved fuel into Chemical Separation.

Chemical Separation

In the Chemical Separation HA Cycle, the UNF dissolved in nitric acid is contacted, using pulsed columns, with an immiscible kerosene solvent containing tri-n-butyl phosphate (TBP). This extracts all the uranium (U) and plutonium (Pu), and a portion of the neptunium (Np), but leaves all the fission products (FPs) and other transuranics within the aqueous acid stream. This stream is, by definition under US regulations, a high level (HL) or highly active (HA) waste stream (HLW or HAW) and it is evaporated to reduce its volume and sent to HA Waste Vitrification. The solvent stream is then contacted, again in pulsed columns, with aqueous chemical reducing agents, causing the Pu, Np and some of the U

to transfer from solvent to aqueous, thus giving two product streams – one with bulk U, the other with mixed U, Pu and Np. The bulk U stream flows to the UP Cycle, where further solvent extraction cycles, this time using mixer settlers, are used to purify the U from residual fission products. The mixed Pu, Np and U stream flows to the TP Cycle where a further pulsed column based solvent extraction cycle is used the purify the Pu/U. The Np can, at this stage, optionally be removed as waste and combined with the HA waste stream, or allowed to continue with the Pu and U. This is achieved by adjustments to the chemical redox conditions. For the current waste estimate, the Np was assumed to be routed to the HLW for vitrification.

The waste streams from the UP and TP cycles contain the residual fission products and the Np. Because "salt-free" reagents are used throughout the HA, UP and TP Cycles, and because the wastes are kept in the acid state, the waste streams from all three cycles can be combined and sent via Salt-Free Evaporation direct to HA Waste Vitrification. There is no need, as there is for the PUREX HL waste from the Hanford and Savannah River waste tanks, for a pretreatment step to remove sodium prior to vitrification. There are four other waste streams from Chemical Separation:

- Condensate from the HA Waste evaporation. This is also salt free and flows to Liquid Effluent Treatment, which is described in a later section.
- Salt Wastes. These wastes arise from the sodium hydroxide and sodium carbonate solutions used to wash and thus re-condition the solvent that recycles though the solvent extraction cycles. These wastes are also treated in separate part of Liquid Effluent Treatment.
- Purged solvent from the HA, UP and TP Cycles. Periodic introduction of fresh kerosene-TBP solvent is required to limit the long term build up of unwashable solvent degradation products. The solvent that must be purged to keep the overall volume constant is destroyed by pyrolysis and the ash from this is consigned, via the Balance of Plant processes, as a Class A waste.
- Hot cell ventilation HEPA filters. These are compacted and consigned as Class A or Class C waste depending on their radioactivity.

Ad-hoc and maintenance wastes from Chemical Separation and Head End Dissolution and Feed Clarification will be very limited, because of the extensive use of Passive Secure Cells [9] instead of the typical US "canyons" to enclose and shield the highly radioactive processes. PSCs (sometimes known as 'Black Cells") and the equipment within them are designed to need no maintenance through the life of the plant, by using equipment with no moving parts or with the maintainable parts of them located outside the radioactive enclosure. Thus moving mechanical equipment that will need maintenance or replacement during the life of the plant is kept inactive and can be consigned to normal commercial non-radioactive waste systems. Canyons, by contrast, are designed to allow the remote disconnection and repair/replacement of equipment (pumps, valves etc) that are in direct contact with radioactive process liquids and so these become radioactively contaminated and must be disposed as radioactive waste.

Product Solidification

Product solidification takes the U nitrate and U-Pu nitrate solutions that come from Chemical Separation and converts them respectively to uranium trioxide and mixed uranium dioxide/plutonium dioxide.

The bulk uranyl nitrate solution is evaporated to high concentration (1000 gU/L) and converted to uranium trioxide powder by Thermal Denitration, using a fluidized bed process in which the highly concentrated uranyl nitrate is sprayed into a tower up which hot air is flowing. The resulting free-flowing UO_3 powder is packaged into stainless steel drums and is then available either for re-enrichment before re-use as fuel in LWRs, or direct use without re-enrichment as mixed oxide (U+Pu) fuel in both LWRs and Fast Reactors.

The mixed U/Pu nitrate solution is evaporated and chemical valence-conditioned to ensure all plutonium is in the +4 state, the uranium present remaining in the +6 state. Uranium in the +4 state is separately produced as uranous nitrate by chemically reducing some of the bulk uranyl nitrate product stream from Chemical Separation. This is then added at a mass either equal to the mass of U(6) already present, or

adjusted so that the U-Pu ratio is exactly that required for the MOX fuel planned to be made from the product oxide powder. This mixture is reacted with oxalic acid to precipitate the U(4) and Pu(4) as uranium and plutonium oxalates. The U(6) is not precipitated and is recycled to the feed into Chemical Separation. The oxalate precipitates are then calcined to produce mixed Pu and U dioxide powder which is packaged into sealed stainless steel containers for dispatch to MOX fuel fabrication.

The only process wastes from Product Solidification are the condensed evaporator overheads, and the off-gas scrubber effluent, which are both routed to Liquid Waste Treatment. Other wastes include hot cell ventilation and thermal denitration off-gas HEPA filters which are compacted and consigned as Class A or Class C waste depending on their radioactivity, plus maintenance and cleanup wastes.

HA Waste Vitrification

The HA waste stream to HA Vitrification comprises the evaporated aqueous waste streams from the HA, UP and TP solvent extraction cycles in Chemical Separation, plus the undissolved fission products from Fuel Dissolution and Feed Clarification. Vitrification is achieved using a Joule Heated Ceramic Melter (JHCM) of the same design as that currently used in the Defense Waste Processing Facility (DWPF) at the Savannah River, U.S.A. site and planned to be used at the Hanford Waste Treatment Plant (WTP). The JHCM is well proven both in DWPF and before that at the Savannah River M Area and at the West Valley facility in New York State. In this type of melter the liquid slurry containing the waste plus the glass formers are fed continuously and directly into the melter where they form a cold cap that progressively melts into the melt pool. This differs from European vitrification systems which have a separate calciner and thus feed powder into the melter. The JHCM is operated continuously with the product molten glass being periodically discharged and poured into standard 600 mm (2 foot) diameter by 4500 mm (15 foot) long stainless steel canisters which are welded shut. In the GNEP work, for the NUEX process, it was planned that these canisters would be consigned to an engineered, passively-cooled store for a period of up to 100 years. This would allow the cesium and strontium activity to decay significantly thus either reducing the heat output of each canister or allowing more waste to be incorporated into the glass. This allows closer packing within the ultimate geologic repository or allows fewer glass canisters to be produced per unit of UNF reprocessed. However, for the present study this delay storage is only considered to be an option.

There are two other waste streams from HA Waste Vitrification:
- Liquid effluent from the melter off-gas scrubbers. This is sent to Liquid Effluent Treatment.
- Hot cell ventilation HEPA filters. These are compacted and consigned as Class A or Class C waste depending on the radioactivity.

Ad-hoc and maintenance wastes from HA Waste Vitrification will again be very limited because of the use of PSCs. An exception to this is the periodic, but now proven infrequent by DWPF experience, need to replace failed melters.

Liquid Effluent Treatment

The HA salt-free waste stream from the HA Cycle in Chemical Separation has nitric acid and water recovered from it during the HA Evaporation step. The lower activity salt-free waste streams from the UP and TP Cycles also have nitric acid and water recovered from them during the Salt-Free Evaporation step. The recovered nitric acid is recycled to the Fuel Dissolution step, thus limiting the amount of fresh nitric acid required to feed the process. The recovered water is combined with all other salt-free condensates and effluents and cleaned of any residual radioactive components by treating with ion exchange materials. This stream is then recycled to the Reagent Gallery where all reagents to Head End and Chemical Plants are made up.

The tritium that comes into Chemical Separation from the Head End is thus recycled in both the recovered acid and recovered water. The water recycle requires a periodic volume purge because some fresh water is inevitably admitted to the system via the new reagent feeds. This purge is encapsulated in a

cement grout and is then consigned as Class A waste. At steady state operation it will contain the same daily amount of tritium as is present in the daily feed of UNF to the process. The tritium content of the purged water is dilute and this gives rise to quite large volumes of purge water to be grouted and disposed as a Class A waste (~12 m^3 of tritiated water per day for a reprocessing plant capacity of 800 tonnes UNF per day). An option exists to concentrate the tritium via the Combined Electrolysis Catalytic Exchange (CECE) process, developed by Atomic Energy Canada Ltd (AECL) at its Chalk River Laboratories [10]. The products from this process are a large volume non-tritiated water stream than can be disposed of as inactive waste, and a small volume, concentrated tritiated water stream (only about 20 L per day) that is cement grout encapsulated and, because it is now highly concentrated in H-3 (typically ~200 Ci/L), it must be disposed as a Class B waste. This tritium concentration option was selected for the present waste estimate.

The salt wastes that arise from solvent washing in Chemical Separation and from certain alkaline off-gas scrubbing steps are evaporated in the Salt Evaporator. The salt-free overheads from this evaporator are recycled to salt-free effluent treatment, while the salt-containing concentrate is sent to a separate cement grouting line and consigned as a Class C waste.

Extensive use is again made of PSCs in Liquid Effluent Treatment so very little radioactively contaminated ad-hoc and maintenance waste is produced.

METHOD OF ESTIMATING WASTE VOLUMES AND TYPES

Summary of Assumptions Underlying the Waste Estimating Process
In estimating the waste volumes and their classes a number of underlying assumptions were made:
- Waste water discharged from the reprocessing facility must meet US Environmental Protection Agency (EPA) drinking water standards.
- Intentional blending of wastes solely to change their waste classifications is not allowed.
- Packaging for GTCC wastes is assumed to be the same as used for remote and contact handled transuranic (TRU) defense waste currently being consigned to the Waste Isolation Pilot Plant (WIPP).
- Vitrified fission product waste (the HA waste from Head End and Chemical Separation) is packaged in 610 mm (2 foot) diameter by 4572 mm (15 foot) tall stainless steel canisters, the standard size that will be used for the vitrified product canisters of defense HL waste from the Hanford Waste Treatment plant. There is an assumed heat load limit at the time of production of 14,000 watts per canister. For 5 year cooled UNF, this was found to be the limiting factor; for the 50 year cooled fuel the limiting factor was waste incorporation capacity into the glass, resulting in a smaller number of canisters per tonne of 50 year cooled fuel reprocessed.
- Carbon-14, krypton-85, iodine-129, tritium and ruthenium-106 must all be captured from off-gas streams and treated for disposal. The first three of these are all captured in the Head End dissolver off-gas treatment processes, tritium is captured in the water purge from Chemical Separation and ruthenium is captured in the vitrification melter off-gas treatment processes and returned to the vitrified HA waste.
- Uranium recovered from the reprocessing must be sufficiently free from fission products and neutron absorbing U isotopes to allow it to be re-enriched and used as reactor fuel. Compliance with the American Society for Testing and Materials (ASTM) standard for recycled uranium ensures this and is routinely and readily achieved by reprocessing plants based on PUREX technology.
- The assumed capacity of the reprocessing facility is 800 tonnes per year and this was used as the basis for estimating wastes arisings. The NUEX process was designed to provide a daily throughput of 5 tonnes per day (and potentially a higher than 800 tonnes/year capacity). For 800 tonnes/year, this translates into 160 operating days per year and this is the basis for estimating process waste amounts that are dependent on the number of operating days.
- Waste estimates are based on normal operation of the facility. The effects of off-normal operations and process upsets were not assessed at this time.

Waste Estimating Methodology

The waste estimating process was carried out in two ways, depending whether waste was *Process Waste* (that is arising directly from the operation of the unit processes, dependant on the number of operating days and the amount of UNF processed per operating day) or *Maintenance and Clean-up Wastes* (that is disposal of failed equipment, materials contaminated during maintenance operations, routine change-out of ventilation filters, general housekeeping wastes that are radioactively contaminated etc).

For Process Wastes, the starting point was the dynamic flowsheet model, set up during the GNEP project, using the Aspen Custom Modeler (ACM) suite of software, to model all the unit processes in the NUEX facility. This model was developed using a combination of first principles equilibrium and rate chemistry for the reprocessing processes, and the decontamination factors and reaction extents that have been observed in practice during the operation of THORP at Sellafield in the UK. The complete isotopic compositions of 50 GWD/tonne UNF for both the 5 year cooled and 50 year cooled cases, which were provided by the DOE as Government Furnished Information, were used as inputs to this model. Such data is typically produced by running the ORIGEN code which assesses LWR reactor physics to estimate the extent of conversion of the uranium fuel into the range of uranium and transuranium isotopes and the range of fission products and their isotopes that are found in fresh irradiated nuclear fuel, together with their decay paths with out-of-reactor cooling time. Operation of reprocessing plants, and the chemical and radiochemical analysis of their dissolved fuel, has provided excellent opportunities to check the output from ORIGEN and the equivalent European FISPIN models against actual UNF, and agreement has been very good.

Operation and convergence of the ACM model with these inputs then produces outputs that define the radionuclide content of every product and waste stream from the reprocessing processes. These outputs become the inputs to an Excel spreadsheet based model that was also developed during the GNEP project. This spreadsheet model has as inputs (i) the ACM process and waste stream specifications, (ii) applicable Nuclear Regulatory Commission (NRC) waste form and classification regulations, (iii) US Department of Transport (DOT) radionuclide transport regulations, (iv) specific activities of every radioisotope in the ACM outputs and (v) our proposed range of waste forms and their capacities for the radioisotopes that we intend to use them for. The output from this spreadsheet then provides the base information to allow the construction of the Process Wastes database which provides fields covering (i) Process Area, (ii) Waste Description, (iii) Raw Waste Volume per day, (iv) Processed Waste Volume per day, (v) Waste Encapsulation method, (vi) Waste Packaging and (vii) Waste Classification. A summarized version of this information is provided in Table 1. Except for the HLW vitrified glass waste form, the U.S. Code of Federal Regulations Chapter 10, Part 61 (10CFR61) was used as the primary basis for determining waste classification. It should be noted that 10CFR61 may not be applicable for all these waste forms, and the exact waste form classification will depend upon the regulations in effect at the time the waste is ultimately created.

For Maintenance and Clean-up Wastes, it was recognized that the arisings of these are not generally dependant on UNF processing rates or even the number of processing days per year. For any large reprocessing facility of 800 to 1000 tonnes per year capacity these wastes are likely to arise at a similar rate, and the best indication of the amounts of such waste will be actual operating experience. Therefore the documented experience from the THORP plant, from the start of its hot operation in 1994 to the present time was drawn upon, with the emphasis on later years during periods of steady operation. The THORP data shows overall totals of UK Classification Low, Medium, and High level wastes plus Pu-contaminated wastes generated yearly from the whole of THORP. These waste arisings were appropriately re-classified under the US system, with all Pu-contaminated wasted assumed to be GTCC TRU-contaminated waste and all other wastes assigned as Class A, B or C waste. As noted previously, the use of Passive Secure Cells instead of Canyons as the shielded enclosures in most of the NUEX reprocessing facility does ensure that little or no contaminated failed equipment requires disposal.

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RESULTS OF THE WASTE ESTIMATING PROCESS

The estimated waste amounts from the reprocessing of both 5 year cooled and 50 year cooled 50 GWD/tonne UNF are summarized in Table 1, which is organized by waste class.

The Table shows all process wastes and all maintenance and clean-up wastes as daily (operating day) or yearly amounts as appropriate. Suitable waste containers are proposed for all types of waste except the UNF MPCs, and this then enables the number of containers per year requiring disposal to be estimated. There are no containers specified for MPCs – these are containers themselves and their internal structure prevents compaction. Currently they are therefore shown for disposal as-is, after a single use, but when a reprocessing facility is designed it will be worthwhile assessing recycling these MPCs for further use in transporting UNF. Another option would be transport of 'bare' UNF directly from reactor storage pools to the reprocessing facility using reusable shipping casks, thus not requiring MPCs. However, this would require either a suitably large pool at the reprocessing site to contain all UNF prior to reprocessing, or there would need to be a means provided for canisterizing the UNF so it can be stored in dry casks.

TABLE 1 Summary of Wastes Arising from Reprocessing 50 GWD/tonne UNF at 5 year and 50 Year Out-of-Reactor Cooling

Waste Class (Note 1)	Waste Components	Daily or Yearly Volume (Note 2)		Disposal Container	Containers per Year		Comments
		5 yr cooled	50 yr cooled		5 yr cooled	50 yr cooled	
HLW	HLW & undissolved FPs vitrified with glass formers	1.1 m³/day	588 L/day	600mm by 4500mm (2ft by 15ft) SS canisters	144	77	5yr cooled limited by heat loading, 50 yr cooled limited by incorporation rates
Remote Handled GTCC	Compacted cladding hulls	0.88 m³/day	0.88 m³/day	RH-72B container	158	158	RH-GTCC because of TRU potentially left with the hulls
GTCC	I-129 sorbed on silver mordenite	23 L/day	23 L/day	55 gallon drums	19	19	Final encapsulant not yet defined. Un-encapsulated volume shown
	TRU-bearing maintenance & clean-up	108 m³/year	108 m³/year	110 gallon drums for 50w% of the waste	104	104	Three compacted 55 gallon drums inside one 110 gallon drum
				55 gallon drums for 50w% of the waste	313	313	50w% is non-compactable so is disposed in the 55 gallon drums
Class C	C-14 as calcium carbonate in grout	53 L/day	53 L/day	55 gallon drums	43	43	Class C because of adhering UNF dust
	Salt Concentrate in grout	9.7 m³/day	9.7 m³/day	High Integrity Containers (HIC) - 210 liners	301	301	
	Maintenance & clean-up	87 m³/year	87 m³/year	110 gallon drums for 70w% of waste	127	127	Three compacted 55 gallon drums inside one 110 gallon drum
				55 gallon drums for 30w% of the waste	163	163	30w% is non-compactable so is disposed in the 55 gallon drums

Class B	Tritiated water Purge	23 L/day	23 L/day	55 gallon drum	19	19	Class B for 5 yr cooled UNF - H-3 concentrated by CECE process.
Class A	MPCs	5.45 m³/day	5.45 m³/day	None	80 MPCs	80 MPCs	No compaction possible, so no over-container used
	Ion Exchange media	11.2 m³/day	11.2 m³/day	210 liners (HIC)	331	331	
	Spent solvent pyrolysis ash	541 L/day	541 L/day	110 gallon drum	206	206	
	Maintenance & clean-up	890 m³/year	890 m³/year	110 gallon drum	2140	2140	
Unclassified	Krypton-85	4.76 L/day	4.53 L/day	49 L gas bottle at 2400psig	15.5	14.8	Delay store and release assumed, so not formally a waste

Note 1: Waste classification shown is best assessment at the present time and subject to change
Note 2: Includes containers

Differences between wastes arising from reprocessing 5 year and 50 year cooled UNF

A major conclusion that can be drawn from Table 1 is that there are only quite small differences in wastes arising from reprocessing of 5 year and 50 year cooled fuel. The one major difference is of course the HLW glass, where annual arisings of canisters are almost halved when reprocessing 50 year cooled fuel. This is mainly because, for 5 year cooled fuel, the specified heat load limit of 14,000 watts per canister is limiting, while for 50 year cooled fuel this limit is not reached, even when loading waste up to the chemical limit of 19w% for incorporation into the glass matrix.

The other wastes for which reprocessing 5 and 50 year cooled UNF might be expected to show a difference are the grouted tritium concentrate and the compressed krypton-85. In the case of tritium, reprocessing 5 year cooled fuel releases about 2110 Ci per day of tritium, while for 50 year cooled fuel this figure is only 248 Ci/day. However, the CECE tritium concentration process output cannot practically go below about 20 L per day and this can contain all 2110 Ci of tritium produced from the 5 year cooled fuel. Therefore the lower 248 Ci of tritium is also contained in the same 20 L/day of tritiated water output, and this becomes the 23 L/day shown in Table 1, once cement grouted. It is possible that research or commercial uses will be found for the concentrated tritium product, in which case it could cease to be a waste.

For krypton-85, this decays some 18-fold as the UNF ages between 5 and 50 years. However, Kr-85 makes up only about 4.8% of the mass of krypton in the UNF, the rest consisting of the inactive isotopes Kr-80 through 84 and Kr-86. So, even an 18-fold reduction in Kr-85 as the UNF ages makes only a small difference to the total amount of krypton that is removed by the cryogenic process. Of course, there may be no need to remove and store krypton-85, especially for 50 year or older UNF, and dependent on the regulatory environment at the time of designing the reprocessing facility. In that case the cryogenic process and associated waste storage could be eliminated.

Waste Destinations for Radioactivity in the UNF

The other major point to take from Table 1 is summarized in Table 2. This shows that, for 5 year cooled UNF reprocessing using NUEX, over 97% of all the radioactivity that was present in the incoming UNF goes into the HLW glass. This percentage is lower than the >99% quoted for THORP because in THORP the Kr-85 is discharged to the atmosphere and the tritium is discharged to the ocean. The THORP vitrified HLW glass thus contains over 99% of the remaining radioactivity in the UNF. For NUEX, the 97% is added to by the stored Kr-85, which accounts for another 2.72%, and the grouted tritium, which accounts for a further 0.11%, so this totals 99.93% for these Group 1 wastes. The remaining amount of radioactivity is split between the rest of the process wastes and the maintenance and clean-up wastes. Most of this is taken up by the remaining process wastes - salt concentrate, carbon-14, cladding hulls, and

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iodine-129 (the Group 2 wastes), leaving only relatively trivial amounts of radioactivity (typically <0.001%) to be distributed around the Group 3 wastes, including the MPCs, ion exchange media, solvent pyrolyis ash), and the maintenance and cleanup wastes.

Modern reprocessing facilities do not therefore spread significant radioactivity into numerous waste streams, as is sometimes claimed, but instead concentrate nearly all of it into a robust waste form, suitable for safe long-term storage and disposal, and arguably more suitable for this than UNF assemblies.

TABLE 2 Distribution of Wastes amongst the Waste Streams for 5 year cooled 50 GWD/tonne UNF

Waste	Classification	Radioactivity as % of Total in UNF	Comments
Group 1 Wastes			
HLW Glass	HA Waste	97.0969	
Krypton-85	Unclassified	2.7195	Unclassified because interim stored only
Tritium	Class B	0.1100	Class B because of concentration by CECE process
Sub Total (Group 1)		**99.9264**	
Group 2 Wastes			
Salt Concentrate	Class C	0.0459	
Carbon-14	Class C	0.0177	Class C because of transferred fuel dust
Cladding hulls	RH-GTCC	0.009	RH-GTCC because of suspect TRU content
Iodine-129	GTCC	0.000013	
Sub Total (Group 2)		**0.0726**	
Group 3 Wastes			
TRU-bearing maintenance wastes	GTCC		
Maintenance wastes	Class C		
MPCs	Class A	**0.000987**	
IX Media	Class A		
Solvent Pyrolysis Ash	Class A		
Maintenance wastes	Class A		

Waste Volume Reductions

Reprocessing provides a significant volume reduction from the UNF in an MPC to the HLW glass for the bulk of the radioactivity, as illustrated in Table 3. The volumes quoted in this Table are based on reprocessing the 10 tonnes of 50 GWD/tonne, 5 and 50 year cooled fuel that is contained in one MPC, which has a volume of 10.9 m^3. The figures are thus equivalent to 2 days of processing UNF at the rate of 5 tonnes per day.

It can be seen that, for 50 year cooled fuel, 99.92% of the radioactivity in the UNF contained in the 10.9m³ volume MPC is transferred to the HLW glass, krypton storage bottles and tritium grout drums (Group 1 Wastes), which have a combined volume of 1.28m³. This is a volume reduction of some 8.5-fold from the original MPC, for the waste that must ultimately be stored in a geologic repository. The equivalent volume reduction for 5 year cooled fuel is lower at 4.6-fold, due to the 14,000 watts/canister heat limit imposed, for the current waste estimating exercise, on the vitrified product. This heat limit arises from the requirement to maintain a maximum center line temperature for the glass in the 600 mm (2 foot) diameter canister, with the canister stood in still air. The NUEX process proposed storing smaller diameter standard European sized vitrified product canisters for up to 100 years in a passively cooled engineered store, with air movement provided by natural convection.

Table 3 Waste Volume Changes from Reprocessing 50GWD/tonne, 5 and 50 year cooled UNF
(beginning with an MPC of volume 10.9m³ and containing 10 tonnes of UNF)

Waste Form	Volume Processed/Produced per 2 Days*		Radioactivity as % of that in the UNF	Initial to Final Volume Ratio		Comments
	5 yr Cooled	50 yr cooled		5 yr Cooled	50 yr Cooled	
Group 1 Wastes						
HLW Glass	2.3 m³	1.2 m³	97.1	4.6	8.5	This group gives a volume decrease. Achievable volume decrease limited for 5yr cooled fuel by 14,000 watt/canister limit for HLW glass
Krypton-85	9.5 L	9.1 L	2.7			
Tritium	49 L	49 L	0.11			
Sub Total	**2.35 m³**	**1.28 m³**	**99.92**			
Group 2 Wastes						
Salt Concentrate	19.4 m³	19.4 m³	0.046	0.51	0.51	This group gives a volume increase – caused by the salt concentrate – but radioactive load is small
Carbon-14	112 L	112 L	0.018			
Cladding Hulls	1.76 m³	1.76 m³	0.009			
Iodine -129	49 L	49 L	0.000013			
Sub Total	**19.45 m³**	**19.45 m³**	**0.073**			
Group 3 Wastes						
TRU-bearing maintenance wastes	1.35 m³	1.35 m³	0.000987	0.23	0.23	This group gives a volume increase, dominated by the IX media, the MPCs and the Class A maintenance wastes. But radioactivity associated with this waste is very low indeed
Class C maintenance wastes	1.08 m³	1.08 m³				
MPCs	10.9 m³	10.9 m³				
IX Media	21.3m³	21.3m³				
Solvent Pyrolysis Ash	1.07 m³	1.07 m³				
Class A maintenance wastes	11.13 m³	11.13 m³				
Sub Total	**46.87 m³**	**46.87 m³**	**0.000987**			

* Including containers

The 100 year delay storage was to allow the radioactivity and heat from the relatively short half-life cesium and strontium to decay away, before placement of the canisters within the geologic repository. This allows the waste loading limit of 19w% used for 50 year cooled UNF reprocessing to be applied also to 5 year cooled UNF reprocessing, instead of a heat limit, thus achieving the 8.5 fold volume reduction for 5 year cooled reprocessing. The other two groups of wastes in Table 3 show a volume increase between the volume of the waste forms and the volume of the MPC from which the radioactivity originated. However, the amounts of radioactivity are very small: 0.073% of the original filled MPC radioactivity for the Group 2 Wastes and <0.001% for the Group 3 Wastes.

CONCLUSIONS

This work has provided waste estimates from reprocessing of 50 GWD/tonne, 5 year and 50 year cooled UNF based on an initially defined set of assumptions, established to aid in understanding the differences in waste estimates that have hitherto been apparent. The work has shown that, with a modern fourth generation reprocessing plant such a NUEX, which has been designed to limit the production and volume of wastes, it is possible to get the vast majority (>99.9%) of the radioactivity in the UNF into waste forms that occupy only about one eighth of the volume of the original UNF assemblies. Over 97% of the radioactivity is encapsulated in the HA waste vitrified product, a robust waste form arguably much more suitable for consignment to a geologic repository than UNF assemblies that were not designed for this purpose.

For NUEX, it was proposed to delay-store the smaller diameter European standard HA waste vitrified product canisters from 5 year cooled UNF reprocessing in a passively cooled engineered store, so as to allow the cesium and strontium to decay. This would potentially allow the heat load limit for these canisters to be raised so that the chemical weights incorporation limit becomes the controlling factor, as it is with 50 year cooled fuel. This would allow the full >8-fold volume reduction to be realized for 5 year cooled, as well as 50 year cooled, UNF reprocessing.

Waste volumes could be further reduced if a number of the current assumptions were modified. Krypton storage could be reduced or eliminated if a decision was made, and approved by the regulators, to allow for acceptable release and dispersal - and this is particularly worthwhile to pursue when reprocessing long (e.g. 50 year) cooled fuel, where the amount of radioactive krypton-85 is a small proportion of the total krypton including all non-radioactive isotopes. Tritium, concentrated by the CECE process up to 105 Ci/L, could potentially be classified as a useful product for energy research or commercial applications rather than a waste. MPCs could be decontaminated and recycled to utilities for further UNF transport rather than consigning them to waste after only one use.

ACKNOWLEDGEMENTS

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