Treatment and Disposal of Process Wastes Arising from the Recycle of Spent Nuclear Fuel in the USA - 9094

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ABSTRACT

A team of industrial organizations led by EnergySolutions has been engaged in conceptual design studies as part of the US Department of Energy's Global Nuclear Energy Partnership. These conceptual design studies consider aqueous recycling of used nuclear fuel (UNF) to provide products of uranium dioxide and uranium-plutonium-neptunium dioxide, which will be recycled in thermal reactors. Americium and curium are also separated and formed into targets for transmutation. This paper describes the process wastes arising from EnergySolutions' aqueous-based process for recycling UNF and how they can be treated and dispositioned, within the context of the US regulatory system. The EnergySolutions' approach for managing the wastes arising from recycling UNF in the USA is compared and contrasted with that adopted at Sellafield, in the UK. The approach utilized in the UK at the Thermal Oxide Reprocessing Plant reflects best environmental practice based on the technology available at the time of plant commissioning and a requirement for an integrated waste management solution, whereas that proposed for the USA is based on satisfying the EPA and NRC environmental discharge and waste management regulations contained in the Code of Federal Regulations.

INTRODUCTION

As part of President Bush's Advanced Energy Initiative, the Global Nuclear Energy Partnership (GNEP) seeks to develop worldwide consensus on enabling expanded use of economical, carbon-free nuclear energy to meet growing electricity demand. This will use a nuclear fuel cycle that enhances energy security, while promoting non-proliferation. It will achieve its goal by having nations with secure, advanced nuclear capabilities provide a service of fresh fuel supply and used fuel recovery to other nations who agree to employ nuclear energy for only power generation. The closed fuel cycle envisioned by GNEP requires deployment of technologies that enable recycling and consumption of some long-lived radioactive waste. The benefits of GNEP include:

- Provision of abundant energy without generating carbon emissions or greenhouse gases.
- Waste minimization and reduction in proliferation concerns by recycling used nuclear fuel (UNF).
- Safe and secure deployment of nuclear power by developing nations to meet their energy needs.
- Assurance of maximum energy recovery from still-valuable UNF to limit the number of required United States (US) geologic waste repositories to one for the remainder of this century.

The US Department of Energy (DOE) selected the team led by EnergySolutions to prepare a Technology Roadmap, Conceptual Design Study and Business Plan for a Consolidated Fuel Treatment Center (CFTC) and Advanced Recycle Reactor. DOE will use the information and recommendations provided by EnergySolutions as well as other data and analyses, to evaluate the development and deployment of GNEP activities and to inform decision making on the path forward for GNEP. This paper focuses on the treatment and disposal of process wastes arising from EnergySolutions' process for recycling UNF in the

USA and compares this strategy with that practiced in the United Kingdom (UK) with reference to the Thermal Oxide Reprocessing Plant (THORP) at the Sellafield site in northwest England.

The EnergySolutions team comprises organizations with international expertise in every part of the nuclear fuel cycle. Technology providers in the EnergySolutions team included Westinghouse Electric Company, Toshiba Corporation, Nuclear Fuel Services and Atomic Energy of Canada Limited (AECL). Particularly relevant to this paper, the UK's National Nuclear Laboratory (NNL, formerly Nexia Solutions) is another team partner, which as part of British Nuclear Fuels (who, at the time, operated the Sellafield Site) performed the original development work for THORP. They provided technical support and input to the commissioning of THORP which went into hot operation in 1994 and since then they have continued to be a provider of technical support to its operation.

ENERGYSOLUTIONS' PROCESS FOR RECYCLING UNF

Drivers for Radionuclide Management

The purpose of the CFTC is to produce actinide products that can be recycled for energy production and transmutation while generating a small volume of immobilized high level waste (IHLW) that optimizes the disposal capacity in the National Geologic Repository, Yucca Mountain (YM). Given the purpose of GNEP, EnergySolutions has identified the following radionuclides which need to be optimally managed within the CFTC:

- Volatile fission products (I-129, Kr-85) need to be separated from the Head End processes off-gas before aerial discharge in compliance with 40CFR190.10. Our analyses indicate that I-129 and Kr-85 overall plant decontamination factors of approximately 200 and 5, respectively, will be required for 5-year cooled UNF. C-14 is also considered for removal to satisfy total radiological release limits.
- Tritium, in the form of tritiated water, needs to be managed to meet our objective of no radioactively contaminated liquid discharges to the environment.
- Uranium should be recovered and made available for recycling as fuel and because it is the greatest contributor (>90%) to the volume of IHLW.
- Heat-generating fission products (Cs-137, Sr-90) and actinides (Pu-238, Am-241, Cm-244) need to be dealt with because they otherwise limit the quantity of IHLW that can be emplaced in YM.
- Long-lived radionuclides contributing most to the radiation dose to future populations from IHLW (Tc-99, I-129, Np-237) need to be dealt with.
- Plutonium should be recovered because it is a valuable resource for energy production. However, the desire to minimize proliferation risks means that no pure plutonium must be separated.

EnergySolutions' Process for Recycling UNF and Identification of Process Wastes

EnergySolutions' overall process for recycling UNF is based on optimally managing the above listed radionuclides. The total process is centered on an aqueous separations portion termed NUEX and is illustrated in Fig. 1.

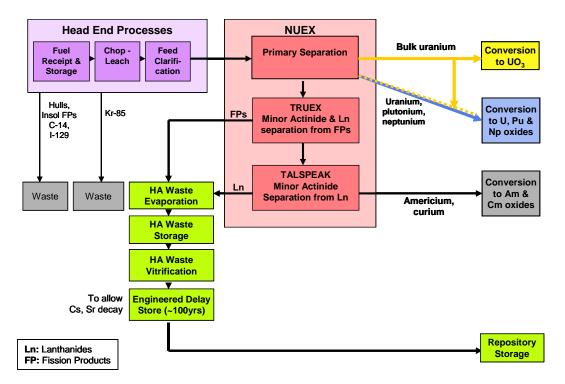


Fig. 1. Overview of EnergySolutions' NUEX-based process for recycling UNF

EnergySolutions' NUEX-based process for recycling UNF is a development of the aqueous process successfully deployed in THORP. EnergySolutions has tailored commercially deployed flowsheets to derive the NUEX flowsheet but the modifications are predominantly chemical in nature such that they have no configurational impact, so that the processing equipment is identical to that already industrially proven in THORP. Therefore, facility design can immediately commence and is not predicated on performing research and development (R&D) *a priori*.

The Head End processes include receipt and storage, UNF chopping and leaching in hot, concentrated nitric acid and leach solution clarification from undissolved fission products, which are very similar to those employed in the THORP Head End. Solid process wastes arising from the NUEX Head End include the fuel cladding hulls and sub-assembly end appendages and insoluble fission products. As in THORP, C-14, in the form of carbon dioxide, is scrubbed from the dissolver off-gas with sodium hydroxide to form a solution of sodium carbonate. Addition of barium nitrate to the spent solution leads to the precipitation of barium carbonate, which is concentrated by decantation. On THORP, I-129 is also removed from the dissolver off-gas by scrubbing with sodium hydroxide. Some I-129 is entrained with the C-14 slurry but the majority remains in solution and is discharged to sea for isotopic dilution. To satisfy US environmental regulations for aerial and wastewater discharges, the EnergySolutions' NUEX approach includes separating the I-129 by adsorption on silver mordenite prior to alkaline scrubbing. No Kr-85 is separated in THORP and it is aerially discharged. To satisfy the current aerial discharge regulations, EnergySolutions proposes to separate Kr-85 by cryogenic distillation after purifying the off-gas of contaminants that might otherwise cause operational problems during distillation.

The NUEX suite of aqueous separations includes three major portions. In primary separation, the uranium, plutonium and neptunium are separated from the remaining transuranics (TRUs) and fission products by solvent extraction processes based on the extractant tri-butyl phosphate (TBP) in a hydrocarbon diluent (such as odorless kerosene, OK, used on THORP). In a second stage of primary separation the mixed uranium, plutonium and uranium stream is further separated into a bulk uranium

product stream and a product stream containing all of the plutonium and neptunium plus a residual amount of uranium at least equal to the mass of plutonium. The presence of residual uranium reduces the proliferation risks associated with plutonium. These two product streams are further purified by TBP-based solvent extraction. Most notably, technetium is separated from the bulk uranium product during its purification by a high acid scrub. The bulk uranium and uranium-plutonium-neptunium products are converted to oxide powders for subsequent fabrication into fuel or potential disposal in the case of bulk uranium. In comparison, THORP also uses TBP/OK but only to extract the uranium and plutonium, which are then completely separated from each other and the products purified before conversion to oxide powders.

In NUEX, americium and curium are separated from the remaining fission products by a further two processes developed by the US National Laboratories:

- The TRU Extraction (TRUEX) process is based on the extractant octyl(phenyl)-N,Ndiisobutylcarbamoyl methylphosphine oxide in a hydrocarbon diluent with TBP added as a phase modifier. The TRUEX solvent extracts the americium, curium and lanthanide fission products leaving the remaining fission products in the aqueous phase, which is vitrified. The vitrified product is decay-stored to allow the radioactivity and thus heat output from the Cs-137 and Sr-90 to diminish before placement in the YM repository. This allows closer packing of the vitrified waste in the repository, thereby allowing increased volume capacity.
- The Trivalent Actinide Lanthanide Separation by Phosphorous Extractants and Aqueous Komplexants (TALSPEAK) process separates the americium and curium from the lanthanide fission products. The lanthanides are added to the fission product waste from TRUEX before vitrification. TALSPEAK uses diethylenetriamine-N,N,N',N",N"-pentaacetic acid to complex the TRUs in a lactic acid buffered aqueous solution while the lanthanide fission products are extracted using di(2-ethylhexyl)phosphoric acid diluted in a hydrocarbon diluent.

The separated americium and curium are either converted to oxide powder for interim storage prior to fabrication into targets or immediately formed into targets. The targets will be irradiated in CANDU heavy water thermal reactors or fast reactors when available to transmute the TRUs to short-lived radionuclides. Removal of the americium from the fission product waste avoids placing a long term heat emitter in the YM repository, again allowing closer packing of the waste. Neptunium, americium and curium are not separated in THORP and instead are vitrified with the fission products.

Several secondary process wastes arise in the NUEX and THORP flowsheets. One is generated from the alkaline washing of TBP/OK and TRUEX and TALSPEAK solvents to remove radiolytic and hydrolytic acidic degradation products and facilitate solvent recycle. Solvent washing uses solutions of sodium carbonate and sodium hydroxide that give rise to a salt waste stream. An organic process waste arises from the small fraction of the solvent that is continuously purged to prevent the accumulation of non-acidic degradation products. Various aqueous nitric acid streams arise as process wastes throughout NUEX and THORP.

PROCESS WASTE TREATMENT, IMMOBILIZATION AND DISPOSAL

Bases of US and UK Approaches

Basis for US Approach

Discharge of radioactive material to the environment is regulated in the USA according to a number of regulations delineated in the Code of Federal Regulations and developed by the Environmental Protection Agency (EPA) and the Nuclear Regulatory Commission. These regulations are largely based on limiting radiation dose to individual members of the public from air and liquid emissions (10CFR20 and

40CFR190.10). One exception is 40CFR190.10b developed by the EPA, which promulgates limits on the quantities of Kr-85, I-129 and TRUs that can be released to the environment.

Licensing of radioactive low level waste (LLW) disposal in the USA is regulated according to 10CFR61. LLW is classified based on the concentration of specific radionuclides in the waste and will fall into one of four classes termed A, B, C and Greater Than Class C (GTCC). Wastes classed as A, B or C are appropriate for near-surface disposal in order of increasing environmental protection. A dual regulatory framework exists for low level mixed waste (LLMW) (i.e. radioactive and hazardous waste) with EPA or authorized states regulating the chemically hazardous portion according to the Resource Conservation and Recovery Act. The NRC or agreement states regulate the radioactive portion of LLMW according to the Atomic Energy Act. A disposal method for GTCC LLW is yet to be determined and the DOE is currently responsible for its disposal. HLW is defined in 10CFR60 and it must be dispositioned in a geologic repository (10CFR60 and 10CFR63).

EnergySolutions has selected technologies and management approaches for the wastes arising from its proposed process for recycling UNF based on satisfying these regulations and taking account of technical and cost factors. EnergySolutions acknowledges significant technical risk and cost associated with satisfying some regulations but is also cognizant of its responsibility to protect the environment as mandated by current regulations. Therefore, we will continue to design for satisfying all US federal regulations while engaging with regulators to re-assess the regulations taking account of factors such as cost, public and worker radiological exposure and technical risk in the context of recycling UNF in the 21st century.

Basis for UK Approach

As a general point, the waste treatment, immobilization and disposal approach used at Sellafield for the wastes arising from THORP utilizes the best environmental practice based on the technology available at the time of plant commissioning in the early 1990s. The background to this approach is that nuclear operators in the UK are required to apply Best Practicable Means (BPM) to control and minimize radioactive discharges [1]. This is one of the principal factors considered by the UK Environment Agency (EA) when issuing discharge authorizations under the Radioactive Substances Act 1993. The definition of BPM is

BPM can be interpreted as that level of management and engineering control that minimises, as far as practicable, the release of radioactivity to the environment whilst taking into account a wider range of factors, including operational safety, technological status, social and environmental factors and cost effectiveness

Therefore BPM requires consideration of a broad range of factors in order to identify the best overall approach for a particular activity; factors that require consideration are not limited to the technology used but also include 'how' the operation of the activity is undertaken.

It should be emphasized here the concept of BPM cannot be equated to specific discharge limits, as what comprises best practicable in one situation may not be the same elsewhere; what comprises BPM will vary depending on the specific context under consideration.

In practice BPM for new facilities is implemented through plant design. Consideration of BPM during early design is preferred as this allows key environmental considerations to be integrated within the process and operations at an early stage. This in turn helps to reduce modifications to design fabrication and constructions at later states and should lead to advantages such as

- Avoidance of waste such as
 - operation of mixer settlers in the reprocessing plants at low speeds to avoid the generation of aerosols containing activity
 - Recycling of acids and separation chemicals where practicable
 - Use of multifunctional waste treatment plants, which accept and treat effluents from a number of upstream plants
- Abatement by design by using equipment which is
 - Demonstrably safe
 - o Reliable
 - Adaptable to allow flexibility for changes in effluent composition
- Dispersion
- Through design of discharge stacks for aerial effluents with varying heights to reflect the impact of the low active effluent being discharged and the ventilation flow characteristics

Therefore, in some respects direct comparison of the Sellafield and proposed US recycling plant waste management approaches needs to be tempered by acknowledging the differences in the UK and US regulatory approaches and the expected technology development achieved over what will become thirty years between THORP commissioning and when we would expect to commission the next US UNF recycling plant. Nevertheless, 40CFR190.10 regulating volatile fission product releases from commercial UNF recycling plants has existed since the 1970s and highlights the more restrictive environmental release limits in the USA.

Solid Wastes

The EnergySolutions proposed strategies for treating, immobilizing and disposing the solid wastes arising from its process for recycling UNF in the USA are compared with those currently practiced in the UK in Table I.

Table I. Comparison of Solid Waste Treatment Strategies Proposed by EnergySolutions for Recycling UNF in the USA with those Commercially Practiced in the UK

EnergySolutions' Process for Recycling UNF in the	Commercial Practice for Recycling UNF in the UK	
USA		
Cladding Hulls and End Appendages		
The cladding hulls are compacted, dried, mixed	Hulls and end appendages are placed in stainless	
with end appendages and packaged in containers	steel drums and encapsulated using composite	
for disposal. EnergySolutions projects the TRU	cement for interim above-ground engineered	
and activation product content of the compacted	storage. These products have been assessed by the	
cladding hulls and ends will classify this waste	UK Nuclear Decommissioning Authority	
stream as GTCC LLW. In addition, activation	Radioactive Waste Management Directorate (NDA	
products and residual fission products will require	RWMD) and classified as being suitable for storage	
the packaged waste to be remote handled (RH).	and repository disposal against the current UK	
	Geological Disposal Facility (GDF) requirements.	

EnergySolutions' US Approach

EnergySolutions considered decontaminating the cladding hulls and ends to render this waste Class C LLW but the technology requirements were considered impractical to achieve, particularly given the significant quantity of activation products. There is currently no GTCC LLW repository available for disposing of the cladding hulls and ends waste stream, although YM and the Waste Isolation Pilot Plant

(WIPP) are potential candidates. EnergySolutions assumed a WIPP-like facility as the repository as a planning and design basis since it currently accepts similar DOE RH-TRU waste arising from defense-related activities. EnergySolutions also assumed RH-GTCC LLW would be transported in RH-72B canisters. It is acknowledged that WIPP is not currently licensed to receive GTCC LLW arising from commercial activities and Congressional authorization would be required for it to do so. Although compaction of this waste stream is a means for its reducing volume, using WIPP and the RH-72B canister as planning bases implies little advantage because one becomes restricted by the canister payload weight limit. EnergySolutions envision that during the implementation phase of recycling UNF, a better optimized container and transport system for RH-GTCC LLW will be adopted, thus the current design basis is considered conservative.

Historic UK Approach

Product Evaluation Programme

The approach taken in the UK for the treatment of Intermediate Level Wastes (ILW), which includes UNF recycling wastes such as barium carbonate, THORP hulls and end appendages and ferric hydroxide Enhanced Actinide Recovery Plant (EARP) flocs, is a highly integrated waste management program known as the Product Evaluation Programme (PEP) to produce wasteforms which meet the requirements of the UK NDA RWMD. This program which has been in operation since 1983 is designed to ensure that all wasteforms which are produced meet the requirements for

- Engineered above ground storage
- Transport and emplacement into the proposed GDF
- Compatibility with the overall GDF concept

Wastes from THORP are treated directly after generation, to produce wasteforms which are suitable for engineered storage above ground, awaiting the availability of a GDF.

The program of work performed to produce the data required to underpin wasteform product quality is in a number of phases to assess the treatment options which were available. These phases are described below.

Phase 1 includes:

- Waste characterization ILW is very diverse because of the wide variety of processes from which it arises. Waste characterization is very important in order to produce simulants for treatability studies which accurately reflect the physical and chemical characteristics of the waste
- Selection and development of non-radioactive simulants for these wastes,
- An initial literature review of encapsulation matrices which were potentially suitable. At this stage the matrices considered included:
 - o inorganic cement,
 - o polymers,
 - o bitumen,
 - o polymer modified cement,
 - o glass,
 - o low melting point metals, and
 - o Ceramics.

Based on the results of this initial review; inorganic cement, polymer modified cement and polymers were selected for additional study in Phase 2.

It should be noted that polymers are used for treating a limited range of other wastes in the UK and that the use of polymers and alternative cementitious material for the treatment of legacy wastes is being actively pursued in the UK.

Phase 2 includes limited small-scale trials that were performed to assess the physical and thermal properties/stability of the selected matrices with simulated ILW streams. Radioactive wastes were used where the activity could affect the properties of the wasteform. The data generated was reviewed to assess the likely impact of scaling up to process scale on the properties, and the most promising matrix for further investigation was selected against a number of essential and desirable criteria. Based on data generated during the Phase 2 studies for a number of ILW streams, a decision was made that inorganic cements should be adopted as the reference matrix for encapsulation of all of the fresh ILW streams arising at the Sellafield site.

Additional studies in Phase 3 were performed to generate empirical data on:

- physical properties (voidage, permeability, porosity),
- physical stability (mechanical strength, impact resistance, dimensional stability),
- chemical stability (cement hydration, waste-matrix interactions, chemical degradation),
- thermal stability (product exotherm, fire stability),
- radiation stability, and
- leaching characteristics (rate of release of radionuclides).

The work was performed at both small- and full-scale, according to the data requirements, with the majority of the studies being performed using non-radioactive simulants that replicate the physical and chemical properties of the active wastes. As in Phase 2, active samples were used where the radioactivity could have an effect on the properties of the wasteform. Data on these properties was collected for periods beyond one year in order to provide information on how the wasteform properties develop over time.

In Phase 4 practical trials to develop acceptable formulation limits for each ILW stream were performed. This work assessed the effect of variables such as waste loading and composition on the wasteform properties.

Operational Database Program

Additional work was undertaken to understand how each process variable affects the quality of the final products. This allowed the processes to be fully understood, and the quality of the products to be predicted with a high degree of confidence. Extensive work was performed during inactive commissioning of the cement encapsulation plants to ensure that they were capable of meeting the constraints defined in the development processes. This overall methodology has been used to develop the encapsulation systems for the four ILW encapsulation plants which have been commissioned on Sellafield site.

UK Waste Management Policy

The concept of geological disposal has been the baseline strategy for the disposal of UK ILW for a number of years with the UK having recently completed an extensive public consultation process¹ entitled Managing Radioactive Waste Safely (MRWS). This concluded that geological disposal is the best available approach to the long term management of waste with the interim engineered storage being an integral part of the strategy. The work described in the integrated programme to determine the properties of wasteforms underpins this strategy, providing the data that gives confidence that the treated wastes can meet the performance criteria for this approach.

The long term management of the vitrified higher activity wastes (which in the UK arise from vitrification of liquid highly active wastes only) from recycling operations is integrated within the MRWS approach, with work ongoing to assess the technical issues and options for disposal of vitrified and ILW within a combined geological disposal facility.

Volatile Fission Products

The EnergySolutions proposed strategies for treating, immobilizing and disposing the volatile fission products arising from its process for recycling UNF in the USA are compared with those currently practiced in the UK in Table II.

Table II. Comparison of Treatment Strategies for Volatile Fission Products Proposed by EnergySolutions for Recycling UNF in the USA with those Commercially Practiced in the UK

EnergySolutions' Process for Recycling UNF in the USA	Commercial Practice for Recycling UNF in the UK
I-129	
Iodine-129 is captured by passing the off-gas through a bed of silver mordenite. The loaded silver mordenite is currently not treated further, and is packaged in containers for disposal as GTCC LLW.	A small proportion of the iodine is captured in the C-14 slurry (see below). The majority is discharged to sea compliant with an approved discharge consent from the United Kingdom Environment Agency (UK-EA).
Kr-85	
Kr-85 gas is recovered from the Head End off-gas stream by cryogenic distillation. It is then evaporated and compressed for storage in bottles. The Kr-85 gas is stored in bottles for decay-storage for up to approximately 25 years (to provide a decontamination factor of 5), when it is released to the environment.	Kr-85 is dispersed from the THORP aerial discharge stack under an approved discharge consent from the UK-EA.
C-14	
The C-14 dioxide gas is captured using a sodium hydroxide scrubber followed by reaction with barium or calcium nitrate to precipitate a barium (or calcium) carbonate slurry. The C-14 slurry is concentrated by decantation and evaporation, mixed with the salt wastes and encapsulated in cement for disposal as either Class A or B LLW.	The C-14 dioxide gas is captured using a sodium hydroxide scrubber followed by reaction with barium nitrate to produce a barium carbonate slurry. This is routed to the Wastes Encapsulation Plant (WEP) for cementation. The cemented C-14 is stored awaiting repository disposal against the current Geological Disposal Facility requirements.

EnergySolutions' US Approach

EnergySolutions projects approximately 90% of the I-129 will be released into the dissolver off-gas. The remainder will remain in solution and most probably be released into the vessel off-gas system downstream of head end. I-129 will therefore require removal from both the dissolver and vessel off-gases to satisfy 40CFR190.10. EnergySolutions expects to accomplish I-129 removal with silver mordenite. Silver mordenite adsorption is considered a relatively straightforward process to implement and has a long history of technology development. It is currently being implemented on the Hanford Waste Treatment Plant for treating the melter off-gas and an adsorption process is also being applied at

the Rokkasho Reprocessing Plant to treat the dissolver off-gas. Significant research under the DOE's Advanced Fuel Cycle Initiative is currently being directed at assessing silver mordenite as a waste form for I-129. This work has identified some heat treatment may be required to adequately fixate the I-129. LLW concentration limits for I-129 are very low and so the I-129 loaded silver mordenite is a GTCC LLW.

Cryogenic distillation is probably the most mature technology for separating Kr-85 and considerable pilot-scale work was completed in Europe and the USA during the 1970s and 1980s. Nonetheless, there remains significant technology development and demonstration for application today. The process complexity will require construction and operation of a pilot plant to demonstrate the integrated system's operability and maintainability and there also remain technical issues associated with Kr-85 immobilization or decay storage. Pressurized decay-storage of Kr-85 gas could present a safety and regulatory challenge and immobilization of the gas would be a preferred approach. Approaches such as ion sputtering into a metal waste form or fixation in a nano-porous material are promising but are considered too immature to consider at this stage of the project. EnergySolutions acknowledges the significant technical risk and cost associated with Kr-85 separation but is also cognizant of its responsibility to protect the environment as mandated by current regulations. Therefore, we will continue to design for Kr-85 separation while engaging with regulators to re-assess the regulations taking account of factors such as cost, public and worker radiological exposure and technical risk in the context of recycling UNF in the 21st century.

C-14 separation, immobilization and disposal methods are more mature and low-risk from a regulatory and technical standpoint and little development is required. Most notably, an alternative precipitating agent, such as calcium, may be considered to avoid the use of barium, which is regulated under the Resource Conservation and Recovery Act. However, immobilization of the C-14 slurry with the salt waste will reduce the barium concentration to a value below that required to pass a Toxicity Characteristic Leach Procedure.

UK Approach

On THORP, I-129 is scrubbed from the dissolver off-gas in the caustic scrubber and discharged to sea compliant with an approved discharge consent from the UK-EA after appropriate treatment. Isotopic dilution in the ocean was considered the BPM for managing this radionuclide at the time of plant commissioning.

Kr-85 abatement in THORP has been the subject of several environmental assessments. A recent UK Government decision [2] authorized the continued release of Kr-85 to air as the best practicable environmental option. This decision was driven by the significant safety issues associated with the operation of a cryogenic gas separation plant and the long lead time in bringing the plant into operation. The decision will be reviewed if the lifetime of THORP is extended for a significant period beyond 2016.

Separation of C-14 from the dissolver off-gas of THORP by caustic scrubbing and precipitation with barium nitrate was considered the BPM for managing this radionuclide at the time of plant commissioning. The C-14 precipitate is encapsulated in cement and stored awaiting disposal in the GDF.

Liquid Wastes

The EnergySolutions proposed strategies for treating, immobilizing and disposing of liquid wastes arising from its process for recycling UNF in the USA are compared with those currently practiced in the UK in Table III.

Table III. Comparison of Treatment Strategies for Liquid Wastes Proposed by EnergySolutions for Recycling UNF in the USA with those Commercially Practiced in the UK

EnergySolutions' Process for Recycling UNF in the	Commercial Practice for Recycling UNF in the UK
USA	
Salt Waste	
The salt waste is concentrated by evaporation to yield a concentrated salt solution and contaminated water. The salt solution is mixed with the C-14 slurry and encapsulated in cement. The contaminated water is further decontaminated of radionuclides and salts by ion exchange and reverse osmosis to facilitate its recycle within the plant. Excess water arises because not all the water can be recycled.	Salt- bearing wastes are neutralized concentrated by evaporation and then decontaminated in the Enhanced Actinide Recovery Plant (EARP), one of the integrated effluent treatment plants on the Sellafield sites. Radionuclides (mainly TRU and technetium) are precipitated by an iron floc process, concentrated using ultrafiltration, and are then cement-encapsulated. The very low level filtrate and evaporator condensate are routed for marine discharge via the Segregated Effluent Treatment Plant (SETP).
Nitric Acid	
Concentrated nitric acid is recovered for recycle by distillation. The contaminated water condensate is further decontaminated of radionuclides and salts by ion exchange and reverse osmosis to facilitate its recycle within the plant.	Acid recovery is performed in the salt free evaporator system which is recycled for use in THORP. The very low level excess water is routed for marine discharge
HLW	
The highly active fission products from NUEX are vitrified in borosilicate glass using one-stage Joule Heated Ceramic melters. The IHLW is decay- stored in an engineered store for approximately 100 years for optimal decay of Cs-137 and Sr-90. The IHLW is then placed in YM for geologic disposal.	The highly active fission products and minor actinides are initially stored in Highly Active Storage Tanks, and then vitrified in borosilicate glass in the two-stage (calcine then vitrify) Waste Vitrification Plant. The vitrified products are stored in engineered above ground stores, while the overall strategy for the vitrified and cemented waste forms disposal is assessed.
Spent Solvent	
Spent solvent is pyrolized and combusted to yield primarily water and carbon dioxide, which are further treated before environmental discharge. An ash gives rise to a solid waste. Treated off-gases are discharged to the environment while the ash is packaged for disposal as Class A LLW.	Spent solvent is routed to the Solvent Treatment Plant (STP), treated by alkaline hydrolysis with caustic soda to break down TBP into three waste streams suitable for treatment and disposal. The kerosene is combusted in a vortex combustor. The TBP breaks down to form sodium dibutyl phosphate which is discharged to sea, where it completely dissolves. An alkaline aqueous phase, which contains most of the residual radioactivity, is treated in EARP before marine discharge.

The THORP salt waste and nitric acid management approaches are considered mature and low-risk for application in the USA. They both give rise to water contaminated with salt and radionuclides, including tritium. Marine discharge of tritiated water and isotopic dilution is an acceptable means of disposing of this waste in the UK in accordance with the approved discharge consent from the UK-EA and is practiced at Sellafield. However, a different approach is required in the USA to comply with current regulations. EnergySolutions approach is to recycle most of the water after ion exchange and reverse osmosis to remove dissolved salt and the radionuclides, except tritium. However, excess water will inevitably arise because, for example, fresh water enters the plant in the concentrated nitric acid reagent necessary to replace that destroyed by thermal denitration of uranyl nitrate solution product and in other processes. Excess water could be sufficiently decontaminated by conventional waste water treatment processes for environmental discharge but for the presence of tritium. Continuing studies are investigating the approaches for optimal economic and technical disposition of tritiated excess water. Options include:

- Immobilization in cement with the C-14 and salt waste to generate Class A or B LLW, for nearsurface disposal.
- De-tritiating the excess water using combined electrolysis and catalytic exchange technology developed by AECL for heavy water de-tritiation so that the excess water is largely discharged to the atmosphere as hydrogen and oxygen gas. The tritium-rich water product would be cemented to generate <1 m³/day Class A LLW appropriate for near-surface burial.

Approaches for high level waste and spent solvent treatment, immobilization and disposal are mature and directly applicable to US application. Spent solvent pyrolysis is practiced in Europe, though not at Sellafield where some unique challenges and the option of marine discharge for liquid wastes made alkaline hydrolysis / combustion more attractive. In contrast, the lack of any liquid wastes from the pyrolysis / combustion approach makes it more attractive for the USA.

CONCLUSIONS

The technologies proposed for the treatment and immobilization of wastes expected to arise from recycling UNF in the US are discussed in the context of the approaches being used for those arising at Sellafield, UK. As in the UK, IHLW and cladding hulls and ends are destined for geologic disposal. Existing US regulations for environmental discharges of I-129 and Kr-85 are more stringent than those in the UK leading to their required separation and immobilization. Water recycle is proposed as the primary means to manage contaminated aqueous wastes in the USA rather than attempting the extensive decontamination required to satisfy regulations for environmental discharge. In contrast, the marine discharge of very lightly contaminated aqueous wastes produced after treatment is an effective management approach for the aqueous wastes arising in the UK. Overall, the approach used at Sellafield for the wastes arising from THORP utilizes best environmental practice based the application of the BPM approach and the technology available at the time of plant commissioning. In addition immobilization of the wastes generated meets the requirements for the proposed GDF in the UK.

REFERENCES

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¹ <u>http://www.corwm.org.uk/Pages/Lnk_pages/about_us.aspx</u>