Downblend and Disposition Remote Handled U-233 Material at the WIPP Facility- #9374

G.M. Polley
Isotek Systems, LLC
P.O. Box 2008, Oak Ridge, TN 37831

ABSTRACT

Isotek was awarded the DOE contract for the downblending and disposition for U-233 material contaminated with TRU isotopes in 2006. The Isotek work scope is the design and construction of a processing facility to dissolve the U-233 material, perform downblending with depleted uranium, concentrate and dry the resulting solution then ship the depleted U-233 waste to WIPP. Upon completion of the processing phase, Isotek will deactivate the facility in preparation for decontamination and decommissioning (D&D).

There are several challenges that will need to be resolved prior to bringing this process operational and ensuring the final result is WIPP compliant.

- The plutonium oxide may not dissolve in nitric acid which may reduce the TRU isotope concentration.
- High radiation levels will require shielded operations and remote handling.
- Radon gas generation can cause facility and equipment contamination issues.

If any of these issues are not adequately resolved prior to startup, the project could have to make costly modifications to the facility and/or the process.

Isotek is determined to find a successful method to resolve these challenges and to do so in a way that is safe, compliant, cost effective, and does not compromise the existing processing and disposition schedule.

INTRODUCTION

ORNL has become the national repository for the U-233 stockpile. Currently the inventory consists of material from Savannah River (SRS), Oak Ridge National Laboratory (ORNL), Rocky Flats (RFTS), Lawrence Livermore National Laboratory (LLNL), and Los Alamos National Laboratory (LANL) in the form of oxides and metals. The initial plan for this material was Thorium extraction for medical isotopes. Specifically, the Thorium 229 extraction would be used to produce Actinium-225 and Bismuth-213 for cancer treatment. The Department of Energy (DOE) had awarded Isotek LLC the contract to perform this work scope but in early 2006 congress changed direction and turned the project over to DOE's Environmental Management Program (EM) to have the material downblended and disposed of at the Waste Isolation Pilot Plant (WIPP). The majority of the U-233 inventory is contaminated with Transuranic (TRU) isotopes and the final waste form will be remote handled due to the high radiation dose rates.

PROJECT SCOPE

The Isotek work scope includes the design and construction of a processing facility to dissolve the U-233 material in a nitric acid solution, downblend with depleted uranium, dry the solution in a Thermal Treatment Unit (TTU) and ship the resulting TRU waste to WIPP for final disposal.

In order to complete this scope, the current facility will be modified to support the material extraction and downblending, a new facility will be constructed for drying and packaging and a storage/loading facility will be modified. Specific elements of the construction and modification activities will include:

- D&D of a contaminated exhaust stack
- D&D of a contaminated manipulator repair shop

- Removal of equipment from the current U-233 storage facility
- Installing a hot cell in the U-233 storage facility
- Building a new HazCat 2 Nuclear facility for drying and packaging
- Installing shielding and a RH-72 canister loading station

These various construction activities will be completed over the next five years with processing beginning in 2012.

PROJECT CHALLENGES

This project has continued to evolve and change since the initial contract award. The U-233 material in storage has not been declared a waste and is still considered product. Discussions have continued on the feasibility of extracting medical isotopes prior to disposition even as the design and engineering has progressed for downblending operations. During these design efforts, it was determined that the original building foundation would not support the planned modifications and a re-design was required. This re-design has included the D&D of an adjacent facility and the construction of a new HazCat-2 nuclear facility for the material drying and waste packaging operations. This facility must also be connected to the main building for transfer of material from the downblend tanks to the feed tanks. In addition to the design and engineering challenges, the amount of characterization data associated with the U-233 inventory is limited and the condition of the storage canisters is not fully known.

Limited Characterization and Material Condition

The analytical data that has been identified by Isotek is limited in regards to quantifying the TRU isotopes and potential hazardous constituents within the U-233 canisters. Due to the high dose rates on the canisters, additional sampling is not currently planned. If the characterization data that was used to plan this project is accurate, all of the material will have adequate transuranic concentrations to qualify for disposal at WIPP. However, this will not be determined until processing begins and samples are pulled from the accountability tanks and an isotopic analysis is performed.

Plutonium Solubility

There are several additional challenges that will need to be resolved prior to bringing this process operational and ensuring the final result is WIPP compliant. A member of the DOE team supporting a design review indicated that since Consolidated Edison Uranium Solidification Program (CEUSP) material was formed at 850^{o}C , the Pu oxide is "high fired" which would make it insoluble in nitric acid. It has been proven that “high fired” pure Pu oxide will not dissolve in nitric acid, but a review of available documentation performed by Nuclear Fuel Services (NFS) provides the following conclusions:

- A study from the Los Alamos National Laboratory indicates that while Pu oxide alone is not readily soluble in nitric acid, Pu oxide up to 25 wt. % in U oxide is readily soluble in nitric acid. [1]. Since there is well under 0.1% Pu oxide in the CEUSP U oxide matrix, the dissolution of the Pu oxide is expected to be rapid.
- Additional supporting data also shows that low levels of Pu oxide in a U oxide matrix dissolves readily. The *Plutonium Handbook* [2] indicates that UO_2 – PuO_2 pellets dissolve “more readily than PuO_2 alone”. On page 579 of this reference, it gives a dissolution rate equation of: [$Rate_{UO2/PuO2} = (Rate_{PuO2})^n (Rate_{UO2})^{1-n}$], *where “n” is the mole fraction of* PuO_2. Thus, for very low concentrations of PuO_2 (as in the case here), the PuO_2 in the U oxide matrix has essentially the same dissolution rate as the UO_2 oxide.
- The trace amount of Pu oxide will be well-dispersed in the U oxide, preventing “crystalline rearrangement, leading to the disappearance of pores as can occur in pure Pu oxide” that exists at

temperatures above 700 °C. [3] The overwhelming mass fraction of U oxide prevents Pu oxide crystalline rearrangement from occurring and the crystalline characteristics of the U oxide will predominate. This finding is consistent with the dissolution rate relationship presented in Finding 2 above.

Based on this information, Isotek is confident that the Pu oxide will dissolve in the uranium matrix and the resulting product will be WIPP Waste Acceptance Criteria (WAC) compliant. In addition to the TRU isotope concentration, a defense determination was performed to ensure that the final product meets the basic criteria for disposal at WIPP.

High Radiation and Contamination Concerns

Another significant challenge to work through is the high dose rate from the feed material and the final product. Each feed canister that is processed in the hot cell will have dose rates up to 300R/hr on contact and the dose rates are expected to be 4R/hr to 12R/hr on contact of the product drums. The 2.6MeV gamma resulting from the decay of Tl-208 is the major contributor to these dose rates which comes from the U-232 decay chain (Table I).

Table I. Uranium-232 Decay Chain

Isotope	Decay Type	Half-Life	Gamma Energy
U-232	α	72 years	
Th-228	α	1.9 year	
Ra-224	α	3.6 day	0.24 MeV
Rn-220	α	55s	0.54 MeV
Po-216	α	0.15s	
Pb-212	β-	10.64h	
Bi-212	α	61s	0.78MeV
Tl-208	β-	3m	2.6 MeV
Pb-208			

The high energy gamma from the Tl-208 presents a significant dose issue that requires shielded overpacks for handling and storing the waste containers and requires remote handling operations for loading waste shipments. Waste is planned to be shipped to WIPP in the 72-B road cask with three waste containers per shipment.

Radon Decay and Contamination

As part of the U-232 decay chain, a continuous source of Rn-220 is created which presents a contamination potential that must be addressed. The current design incorporates a radon decay tube inside the waste containers in order to reduce the radon emissions (Fig 1). The radon decay tube is connected to the NucFil drum filter so the radon gas will have time to decay inside the drum and not be released into the atmosphere. The two main reasons to keep the radon decay inside the drum are to prevent facility and equipment contamination and to minimize the chance of contaminating the 72-B Road Cask during transport.

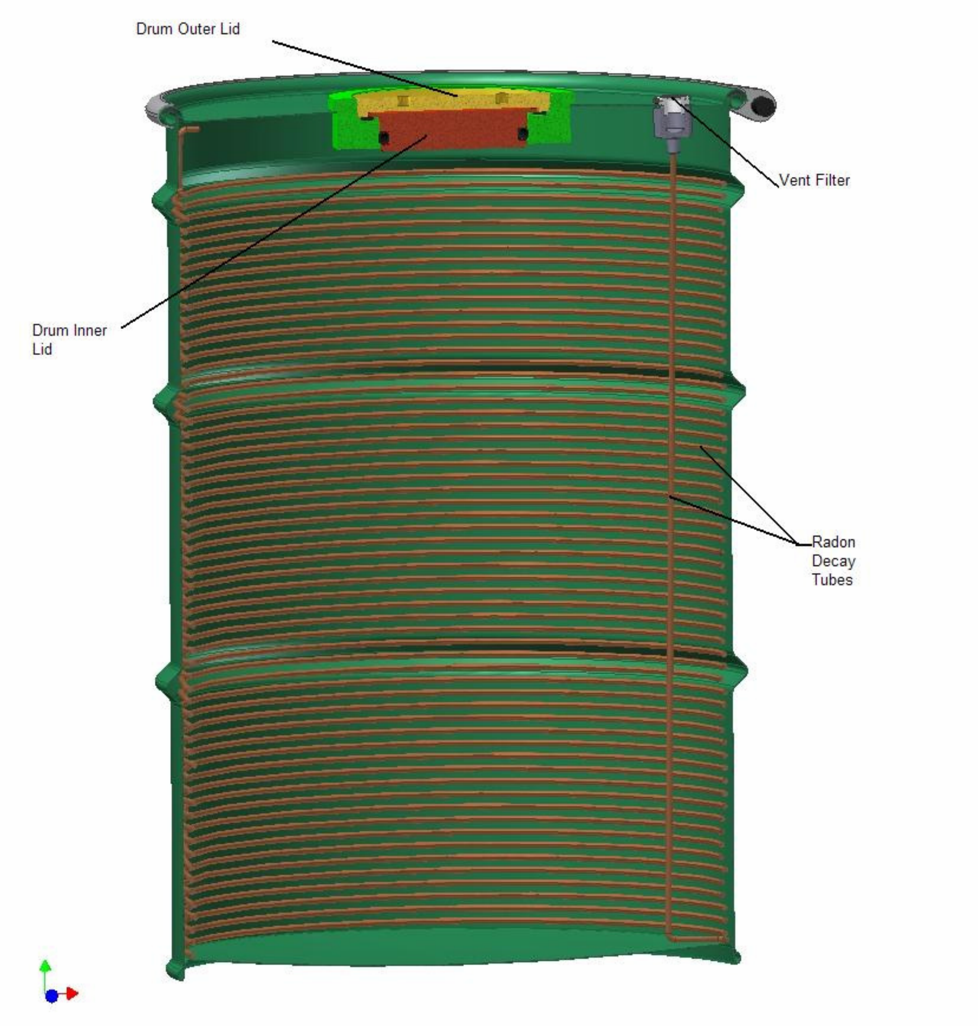

Fig. 1. Waste Container w/Radon Decay Tube

PROCESSING OPERATIONS

The U-233 downblending process is illustrated in Fig 2 and begins by removing material containers from storage and transporting the packages to a hot cell. At the hot cell, the containers will be inspected, the material removed from the canister. Upon completion of the inspection, the material is transferred to the dissolution columns that contain nitric acid. The resulting solution is then transferred to the accountability tanks where a sample will be taken to maintain an accurate inventory of the Special Nuclear Material (SNM). Material from the accountability tanks will be transferred to the down blending tank and combined with depleted Uranyl Nitrate to reduce the U-235 concentration to below 1% in order to meet contractual requirements. The material is transferred to the concentrator feed tank and then on to the Wiped Film Evaporators where the majority of the nitric acid will be removed. The concentrated feed will then be transferred to the Thermal Treatment Units (TTU) for final drying. The dried material flows from the TTU into the waste container. The filled waste containers are transferred to interim storage and prepared for shipment to WIPP.

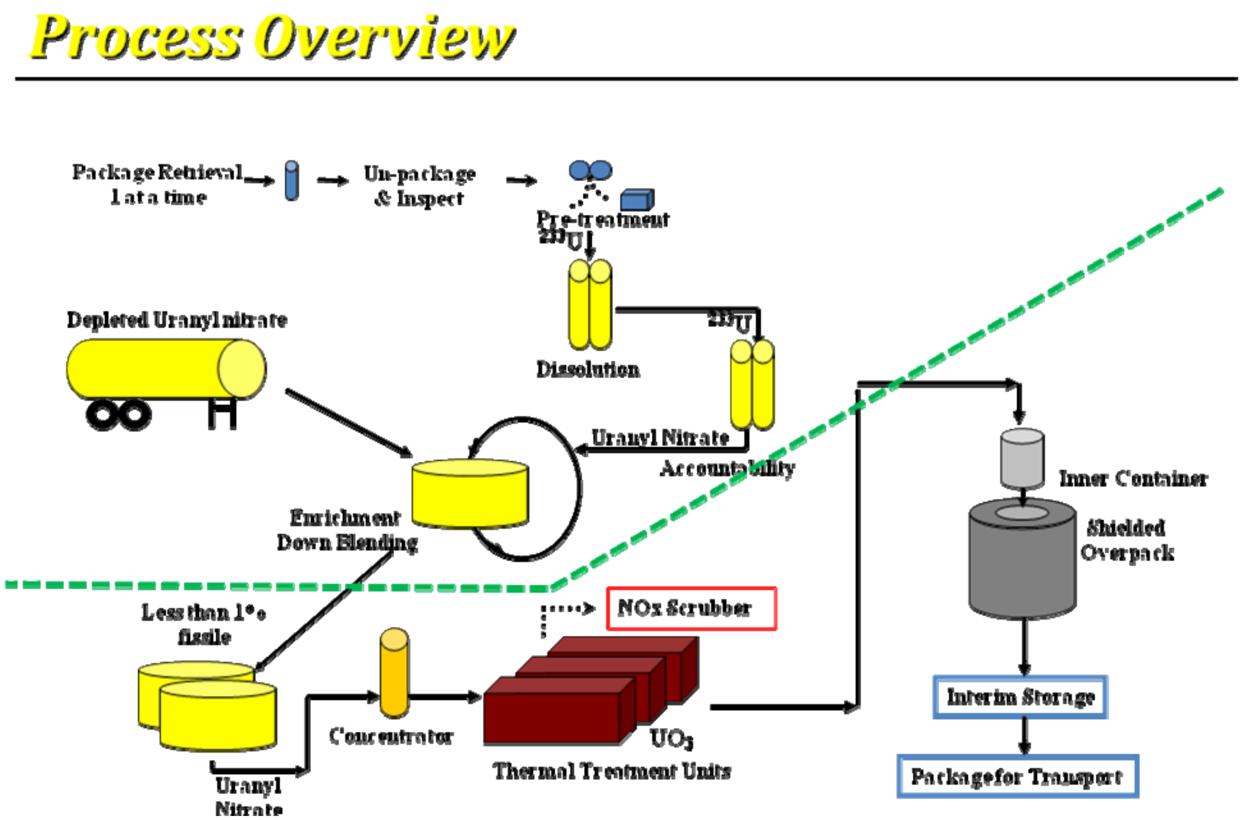

Fig. 2. U-233 Downblending Process Overview

Sampling and Analysis
Sampling will be performed at several points during the process. After dissolution of the material in nitric acid, an accountability sample will be taken to manage the inventory of the enriched U-235 and U-233. A second sample will be taken after downblending to ensure the downblending targets has been reached, <.96% U-235 and to determine the quantity of TRU constituents. The final sample will be taken during waste container filling operations, to verify TRU isotopic concentration and check for the presence of RCRA regulated materials. These sampling evolutions will provide challenges due to the high radiation levels and the contamination control issues. Each 5-7 gram sample will have a dose rate of approximately 54 mR/hr on contact. The samples will be taken remotely and transferred to shielded containers for delivery to the on-site laboratory. Isotek will perform the analysis for all samples and transfer the data from the final product samples to CCP for WIPP required certification.

Packaging, Storage and Transport
The closure of the drum after filling will be performed remotely using a bagless transfer system between the drum and the TTU. The drum will be transferred on conveyors to a shielded area where contamination surveys are taken and the overpack lid is installed. Up to six filled containers can be staged at the processing facility but plans are to transport the filled containers to storage every day. Each drum with the shielded overpack will weigh approximately 8,000 pounds and the truck will carry two containers on each trip. In order minimize the dose to the driver during the transport operations, a six inch steel shield panel will be installed behind the cab of the truck.

At the waste storage facility, each drum will be removed from the overpack and placed in a counting station in order to perform a dose-to-curie measurement to determine the fissile mass of each container. The drums will be returned to the overpack and stored until CCP provides payload configurations based on the characterization data provided. After the payload is determined, Isotek personnel will transfer the drums from the overpacks to a RH-72B canister in preparation for shipping. CCP will then deploy the mobile transportation team to support loading the canisters into the RH-72B road casks for transport to WIPP. The current plans are to make four shipments to WIPP each week during shipping operations.

Facility Turnover

The processing operations will complete in approximately two years and at that point all SNM will be removed from the facilities. Isotek will perform facility shutdown and turnover activities in order to return the site to DOE for final D&D. The facilities and systems will be cleaned and flushed to minimize the amount of contamination and radioactive material remaining in the piping and components. It is expected that the system flushing, decontamination, HEPA filter change out and equipment shutdown will require about six months and the facility will be returned to DOE in early 2015.

REFERENCES

1. DeMuth, Scott F., *Conceptual Design for Separation of Plutonium and Gallium by Solvent Extraction*, LA-UR-97-181, Los Alamos National Laboratory, Technology & Assessment Division.

2. Wick, O.J., *Plutonium Handbook – A Guide to the Technology*, Pacific Northwest Laboratories – Battelle Memorial Institute, page 579.

3. Manchuron-Manard, Xavier, Plutonium Dioxide Particle Properties as a Function of Calcination Temperature, Journal of Alloys and Compounds 225 (1966) 216-224, revised 20 October 1995.