

U.S. Army Corps of Engineers

> F U S R A P

# Niagara Falls Storage Site FUSRAP Site Lewiston, New York

Technical Memorandum: Treatability Study Considerations to Support the Niagara Falls Storage Site Feasibility Study

**Prepared for:**U.S. Army Corps of Engineers
Buffalo District

**Prepared by: Science Applications International Corporation Dublin, Ohio** 

**Contract: DACW49-00-R-0027** 

**November 2002** 

# TECHNICAL MEMORANDUM: TREATABILITY STUDY CONSIDERATIONS TO SUPPORT THE NIAGARA FALLS STORAGE SITE FEASIBILITY STUDY

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# ABBREVIATIONS AND ACRONYMS

**AMCB** African Metals Corporation of Belgium

CAA Clean Air Act

Comprehensive Environmental Response, Compensation, and Liability Act CERCLA

Code of Federal Regulations **CFR** 

Ci Curie

CWA Clean Water Act

DEO Department of Environmental Quality

DOE Department of Energy

DOT Department of Transportation

EMF Electromotive force

EPA **Environmental Protection Agency** Feed Acceptance Criteria and Tests **FACT** 

**FEMP** Fernald Environmental Management Project

FS Feasibility Study

Formerly Utilized Sites Remedial Action Program **FUSRAP** 

GTCC Greater-than-Class C **HDPE** High-density polyethylene **IMC** Inter-modal containers

**IUC** International Uranium Corporation

kg Kilogram

LDR Land Disposal Restrictions

Low-Level Waste LLW

Low Specific Activity II LSA II  $m^2/s$ square meters per second

 $m^3$ cubic meters mg/L milligrams per liter millirem per hour mrem/hr

National Academy of Sciences NAS NFS Nuclear Fuel Services, Inc. Niagara Falls Storage Site NFSS NRC **Nuclear Regulatory Commission** 

NTS Nevada Test Site

Polychlorinated Biphenyl PCB PFD Process flow diagram Perma-Fix of Florida, Inc. PFF

 $Pb^{210}$ Lead - 210

picoCuries per gram pCi/g process knowledge PK parts per billion ppb pounds per square inch psi PVC Polyvinyl chloride

Quality Assurance/Quality Control QA/QC

 $Ra^{226}$ Radium – 226  $Rn^{222}$ Radon - 222

Resource Conservation and Recovery Act RCRA

Radioactive Material Profile RMPR

ROD Record of Decision

RWMS Radio Active Waste Management Sites

Semi-autogenous grinding SAG

SAP Sampling & Analysis Plan

SAIC Science Applications International Corporation

SDWA Safe Drinking Water Act SRS Savannah River Site

TAS Treatability/Amenability Study

TCLP Toxicity Characteristic Leaching Procedure

 $\begin{array}{ccc} \text{Tpd} & \text{tons per day} \\ \text{Th} & \text{Thorium} \\ \text{Th}^{230} & \text{Thorium} - 230 \\ \text{Th}^{232} & \text{Thorium} - 232 \\ \text{U} & \text{Uranium} \\ \text{U}^{238} & \text{Uranium} - 238 \\ \end{array}$ 

UMTRCA Uranium Mill Tailings Radiation Control Act USACE United States Army Corps of Engineers

US EPA United States Environmental Protection Agency

WAC Waste Acceptance Criteria

WSRC Westinghouse Savannah River Company

# **EXECUTIVE SUMMARY**

Science Applications International Corporation (SAIC) is currently conducting a feasibility study (FS) for the Niagara Falls Storage Site (NFSS) under contract with the United States Army Corps of Engineers (USACE) Buffalo District. One of the tasks in the scope of work is to identify technologies that are possible candidates for detailed analysis in the FS and evaluate whether treatability studies are warranted for these technologies. This task focused on the K-65 residues within the Waste Containment Structure (WCS) as they contain the highest activities and were the subject of the National Academy of Sciences (NAS) recommendations. (No specific evaluations were performed as part of this task for other materials at NFSS.)

Treatment of the K-65 residues is being considered in the FS because:

- 1) No offsite disposal facilities can currently accept the K-65 residues for disposal without some form of treatment;
- 2) Department of Transportation (DOT) requirements make transportation without some form of initial treatment, expensive and difficult; and
- 3) The United States Environmental Protection Agency (EPA) Comprehensive Environmental Response, Compensation and Liability Act (CERCLA) regulations provide a statutory requirement to evaluate treatment.

The screening of potential treatment technologies for the K-65 material involved researching literature and other sources to identify potentially applicable technologies. The primary sources for potential treatment technologies were:

- The draft Evaluation Report on Remediation of the NFSS Residues prepared by DOE in 1996 (DOE 1996);
- Technology evaluations performed by Fernald Environmental Management Project (FEMP) for treatment of the Silos 1 and 2 material; and,
- Literature searches of new treatment technology developments.

In 1996 the Department of Energy, Formerly Utilized Sites Remedial Action Program performed a preliminary evaluation of possible removal and treatment alternatives for the high radium concentration residues from the NFSS. As a result of the technology screening, three treatment technologies were recommended for further evaluation: ex-situ chemical separation, ex-situ solidification/stabilization, and ex-situ vitrification (DOE 1996). However, no further evaluation of these technologies was performed by DOE at that time.

The FEMP in Ohio has waste that is currently stored in Silo 1 that is similar to the NFSS K-65 residues. FEMP has performed treatment technology evaluations, conducted bench and pilot scale treatability studies and is finalizing a revised FS that addresses the Silo 1 (and Silo 2) material. Information regarding FEMP's treatability testing and lessons learned from their treatment technology evaluations is presented in Section 2.

The FEMP residues have been declared 11e(2) material not regulated by the Nuclear Regulatory Commission (NRC), and are thus exempt from RCRA regulations. FEMP's primary focus is now on treatment to the extent that it allows cost effective transportation of the waste in compliance with U.S. DOT requirements versus treatment to meet RCRA criteria. The current proposed remedy for the FEMP residues is stabilization/solidification with flyash and/or cement to meet

NFSS – USACE Tech Memo: Treatability Studies for NFSS Page iv DOT requirements, transportation in rolled steel containers, and disposal at Envirocare. The FEMP remedial action is scheduled to be implemented in 2004.

SAIC contacted FEMP representatives to determine whether a sample of the K-65 waste could be obtained from FEMP to use for the NFSS treatability studies. The K-65 residue sample volume that is now available is limited and DOE has indicated their preference for retaining this sample to perform additional studies to refine their remedial design. Therefore, material from FEMP will not reasonably and economically be available for USACE treatability studies until FEMP begins removal of the K-65 residues from the silos for treatment and disposal in approximately two (2) to three (3) years.

Based on a review of the 1996 treatment technology screening performed by DOE, information gathered from the FEMP treatment technology evaluations, and recent technology literature, four technologies are recommended for further evaluation for the NFSS K-65 residues: 1) reclamation (resource/recovery), 2) stabilization, 3) encapsulation, and 4) photodeactivation (transmutation using gamma rays). The first three technologies are conventional and have been used for hazardous and radioactive waste treatment. The fourth technology, transmutation, is an extremely innovative technology that has not been used in the past to treat radioactive waste. Descriptions of the technologies and the justification for their recommendation are presented in Section 2. Abbreviated work plans for treatablity studies to further evaluate these technologies are presented in Section 4.

The conclusions and recommendations from this evaluation of potential treatment technologies and treatability studies are as follows:

- 1) The residues in the WCS derive from ore extraction activities carried out before 1978 and are known as uranium mill tailings as defined in Section 11(e)(2) of the Atomic Energy Act, as amended by the Uranium Mill Tailings Radiation Control Act. Thus under the FUSRAP, USACE has declared the residues 11(e)(2) [pre-1978] material and that the material is exempt from RCRA regulations.
- 2) Due to the fact that treatability data is available from the FEMP treatment evaluations and the unavailability of a residue sample, treatability studies are not required prior to proceeding with the NFSS FS. However, the four technologies (mentioned above) have been identified for further evaluation. Completion of the identified treatability studies would serve, at relatively low cost, to confirm the conclusions being formulated for the FS. In addition, such studies would likely provide valuable design data for full-scale remedial action.
- 3) Due to the fact that the techniques (e.g. slurrying) used to remove the K-65 residues from the WCS will impact any blending and/or treatment operation, removal techniques for the residues should be evaluated prior to performing any treatability studies.
- 4) It is recommended that although the FS can proceed without treatability studies, USACE efforts should begin immediately to establish a cooperative agreement with the DOE for retrieval and use of adequate sample volumes to support the treatment technologies identified in this report and allow the opportunity to obtain additional sample volume should the need arise.

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# 1.0 INTRODUCTION

Science Applications International Corporation (SAIC) is currently conducting a feasibility study (FS) for the Niagara Falls Storage Site (NFSS) under contract with the United States Army Corps of Engineers (USACE) Buffalo District. In support of the FS SAIC was tasked with identifying treatment technologies that are possible candidates for consideration during the detailed analysis of alternatives in the FS, and evaluating whether treatability studies are warranted for these technologies. This task focused on the high activity K-65 residues within the Waste Containment Structure (WCS).

# 1.1 PURPOSE

The purpose of this technical memorandum is to identify candidate treatment technologies for the K-65 residues and present recommendations for treatability studies based on a review of existing documentation [including information from the Fernald Environmental Management Program (FEMP) studies]. These evaluations were focused on the K-65 residues as they contain the highest activities and were the subject of the National Academy of Sciences (NAS) recommendations. (No specific evaluations were performed as part of this task for the other residues and contaminated soils within the WCS.)

It is anticipated that the K-65 residues will require some type of treatment and/or blending with additional materials to reduce the activity of the residues to meet the disposal facility waste acceptance criteria. Treatment and/or blending may also be required along with packaging, shielding, and transportation configurations to meet Department of Transportation regulations.

This document is intended to provide the USACE with sufficient information to determine whether or not it is necessary to pursue the treatability studies in support of the FS alternatives development. It will also give the USACE an opportunity to provide input on the approach and proposed work scope for each technology.

# 1.2 BASIS FOR THE EVALUATION OF TREATMENT TECHNOLOGIES

Treatment of the NFSS residues is being considered in the FS because:

- 1) No offsite disposal facilities can currently accept the K-65 residues for disposal without some form of treatment;
- 2) DOT requirements make transportation without some form of initial treatment, expensive and difficult; and
- 3) The United States Environmental Protection Agency (EPA) Comprehensive Environmental Response, Compensation and Liability Act (CERCLA) regulations provide a statutory requirement to evaluate treatment.

The following items were considered in the selection of potential treatment technologies:

Preference was given to technologies that would address or comply with the NAS
recommendations to develop a program to remove the high level residues from the site
(National Research Council 1995).

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- Preference was given to technologies that could provide for resource recovery of, or minimize the toxicity and mobility of the contaminants in the waste should the waste need to also satisfy RCRA requirements for disposal.
- Volume reduction technologies were considered. However, these technologies, when applied to radioactively contaminated wastes, typically create a more concentrated waste stream (higher activity) which, considering the high activities of the residues, adds another level of risk to waste handling to an already complex situation.

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# 2.0 TECHNOLOGY DESCRIPTION AND JUSTIFICATION FOR SELECTION

The screening of potential treatment technologies for the K-65 material involved researching literature and other sources to identify potentially applicable technologies. The primary sources for potential treatment technologies were:

- Technologies recommended for evaluation by DOE during a 1996 screening effort (DOE 1996);
- Technologies evaluated by FEMP for treatment of the Silos 1 and 2 material; and,
- Literature searches of new treatment technology developments.

# 2.1 TECHNOLOGY SCREENING FOR NFSS RESIDUES

In 1996 the Department of Energy Formerly Utilized Sites Remedial Action Program performed a preliminary evaluation of possible alternatives for removal of the high radium concentration residues from the NFSS. The objectives of this effort were to develop one or more alternatives (including an approach for removal, treatment, transportation and disposal) which were considered technically feasible and which would provide a basis for a 10 year plan budget estimate. The constraints for the evaluation were that the alternatives selected must comply with the NAS recommendations to develop a program to remove the high level residues from the NFSS site and must meet the funding and schedules constraints present at the time of the evaluation (1996).

The alternatives developed and the results of the evaluation are documented in the draft *Evaluation Report on Remediation of the NFSS Residues*, September 1996 (DOE 1996). The treatment technology screening conducted as part of this evaluation is shown in Table 2-1.

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Table 2.1. DOE Screening of Treatment Technologies for the High Level Residues at NFSS<sup>1</sup>

Technology	Pros	Cons	Conclusion				
In-situ Treatment Technologies							
In situ vitrification	Minimizes worker exposure  Reduces handling costs	Requires retrofitting the storage cell  Do not have necessary data  Waste may have high moisture content, thus affecting the amount of energy required  Do not know the final size of the melt	Not recommended for additional evaluation				
		Implementability uncertain due to innovative nature of technology  Geology may not be conducive to this technology					
In situ chemical separation	Minimizes worker exposure  Potential cost savings	Requires retrofitting the storage cell  Engineering issues must be addressed related to use of equipment inside of cell  Do not know the permeability of waste  Implementability uncertain due to innovative nature of technology  Need additional unavailable data (further characterization required)  Percent removal of residues is uncertain  Amount of waste would be increased due to re-slurrying process  Re-slurrying could cause other unknown reactions  Possible compromise in control of	Not recommended for additional evaluation				

	Ex-site	u treatment technologies		
Ex-situ air flotation	Conventional treatment process  The unit (equipment) is relatively inexpensive	Particles in waste may be too fine (a large fraction of the K-65 residues are <37 um in size)  Technology may not be a standalone method	Not recommended for additional evaluation as a stand alone technology May warrant consideration as a pre-treatment method	
Ex situ electrical separation	May be simple and inexpensive	Radon control may be difficult Uncertainty as to whether the materials would separate by electrical gradient  Do not know where the cut-point would be (treatment process parameters unknown)  May have to add a substitute species to the process	Not recommended for additional evaluation	
Ex situ chemical separation	Proven and documented treatment process  Ease in maintaining, monitoring, and chemical control	The process is in research stage – no pilot tests have been performed  May not be able to do anything with the by-product  Would generate significantly more secondary waste  Requires multiple passes  Requires a large plant, increased shielding, and increased costs  Radon control would be a major issue	Recommended carrying forward to next evaluation stage	
Ex situ liquid extraction	100% recovery of uranium if in solution  Would decrease primary waste significantly (this method would achieve the minimum volume of any process)	Would increase volume of secondary waste  There are hazards associated with liquids	Not recommended for additional evaluation	
Ex situ solidification/stabiliz ation	Conventional and simple technology  Low treatment cost  May decrease the radon control problem  Polymers are effective for radon emission reduction	Would increase volume  Would increase transportation and disposal costs  There are questions of long-term stability  Major radon control issue	Recommended carrying forward to next evaluation stage	

Ex situ polyethylene glycol	Suspected to be one of the most efficient methods for uranium removal	Requires changing the suspension fluid	Not recommended for additional evaluation
	WWW.W. Policy W.	Unproven for this waste (sulfates)	
		Requires high salt concentration and temperature and salt variation	
		Results in the generation of an additional waste streams	
Ex situ vitrification	Commercially available	Facility would be expensive	Recommended carrying forward to next evaluation
	Proven technology	All waste would be disposed of offsite	stage. (Recommendation based on the fact that at the time of the evaluation ex situ
	Has been effective with high activity waste		vitrification was the baseline technology for Fernald.)
	A range of costs exist for this technology		
	Could have schedule benefits		
	Process equipment is inexpensive		
Ex situ catalytic extraction	Mobile units may be available	Unproven technology	Not recommended for additional evaluation
	Significant volume	Emissions would be an issue	
	reduction may be achieved	Radon would be an issue	
	Solids would be soluble		
Deep well underground injection	Relatively inexpensive  Permanent solution	May counter the NAS recommendations	Not recommended for additional evaluation
	No transportation, disposal or exposure	Requires heavy-duty hydra- fraction equipment	
	Engineering is feasible	Technology has a history of past failures	
	Geology may be conducive to this method	Public reaction/perception may be negative	
		Geology may not support this technology	

<sup>&</sup>lt;sup>1</sup> Adapted from DOE 1996.

Shading indicates technology that DOE recommended carrying forward to the next evaluation stage.

As a result of the technology screening documented in the draft *Evaluation Report on Remediation of the NFSS Residues* (DOE 1996) 3 treatment technologies were recommended for further evaluation: ex-situ chemical separation, ex-situ solidification/stabilization, and ex-

situ vitrification. However, no further evaluation of these technologies was performed by DOE at that time.

## 2.2 FERNALD TREATMENT EVALUATIONS FOR SILOS 1 AND 2 MATERIALS

As mentioned previously, the FEMP is also addressing the removal, treatment, transportation, and disposal of the K-65 residues (stored in Silos 1 and 2 at FEMP). The proposed remedy for the FEMP K-65 residues is stabilization/solidification with flyash and/or cement to meet DOT requirements, transportation in rolled steel containers, and disposal at a commercial disposal facility (e.g. Envirocare) or NTS. The FEMP remedial action is currently scheduled for implementation starting in 2004.

In 1994 FEMP issued a Record of Decision for OU 4 (which includes Silos 1 and 2). The remedy selected for Silos 1 and 2 outlined in the ROD consisted of removal of the contents of the silos, remediation by vitrification and off-site disposal of the treated material at the Nevada Test Site (NTS). Following approval of the ROD the DOE initiated a joule-heated Vitrification Pilot Plant (VITPP) Treatability Study Program. Technical and operational difficulties encountered during implementation of the VITPP study resulted in schedule delays, cost increases, and culminated with suspension of the VITPP testing. The DOE-FEMP, independent review team, EPA regulators and stakeholders ultimately concluded that the treatment remedy for the Silos 1 and 2 materials should be reevaluated. The EPA directed DOE-FEMP to proceed with development of a supplemental FS/Proposed Plan/and subsequent Record of Decision (ROD) amendment for the Silos 1 and 2 materials. A revised FS (DOE 2000) was prepared to document the reevaluation of the selected treatment remedy for the Silos 1 and 2 materials. The following paragraphs summarize the screening of treatment alternatives presented in the revised FS (DOE 2000).

Several categories of potential treatment technologies identified by FEMP were judged inapplicable for treatment of the Silos 1 and 2 materials and were not included in their technology screening process. The Silos 1 and 2 materials consist primarily of the residue from a solvent extraction process. Therefore, subjecting the material to further solvent extraction does not further reduce the mobility of toxic constituents. Similarly, thermal oxidation of the Silos 1 and 2 material does not provide any appreciable treatment benefit. Thus, the revised FS concluded that solvent extraction and thermal oxidation technologies did not warrant further evaluation. In addition, the revised FS excluded off-site treatment facilities from further evaluation as they lack both the capacity and necessary permits and licenses for treatment of the Silos 1 and 2 material.

The following treatment alternatives were evaluated in the revised FS (DOE 2000):

- Vitrification (joule-heated, cyclone, rotary, and plasma-arc);
- Chemical stabilization (cement and non-cement);
- Polymer phosphate stabilization;
- Encapsulation (ceramic and polymer); and,
- Thermal stabilization.

FEMP concluded that rotary vitrification, ceramic encapsulation, polymer phosphate stabilization, polymer encapsulation, and thermal stabilization were not demonstrated at full-scale on low-level, hazardous, or mixed wastes and therefore, did not warrant further

consideration in the detailed analysis of alternatives. Vitrification (joule-heated and other) and stabilization (cement-based and other) were identified for further evaluation.

FEMP performed bench and pilot scale treatability studies on stabilization and vitrification technologies. Stabilization was selected as the preferred treatment alternative in the revised FS based on its straightforward implementability and safety. Vitrification provided a significant waste volume reduction however, the transportation and disposal cost savings did not justify the high capital cost and complex operations associated with the process.

Lessons learned from the FEMP treatment evaluations and studies that may be applicable to treatment evaluations for the NFSS are summarized below:

- Use of surrogate waste for treatability studies was effective for evaluation of certain
  physical parameters such as dewatering and settling characteristics that are important to
  materials handling considerations. However, use of surrogates to evaluate the vitrification
  technology performance (containment, mobility, and contaminant reduction, etc.) was not
  effective and led to numerous design problems.
- Attention should be given to the retrieval method for the K-65 residues. At FEMP it was concluded that little had been done to ensure that removal efforts would proceed safely, easily, and at the rate anticipated to support the treatment process.
- Additional characterization of the FEMP residues was suggested in order to better understand the characteristics and to assist in developing treatment process recipes.
- The K-65 residue in Silo 1 is very heterogeneous therefore, the treatment process must be sufficiently flexible to adapt to varying contaminant concentrations.
- Regardless of the treatment process selected it was recommended by the FEMP project review team that some form of commercial involvement should be actively pursued rather than in-house design, construction, and operations of a new facility.
- Waste loading for the FEMP stabilization process is primarily controlled by compliance with DOT requirements versus compliance with RCRA criteria.

# 2.3 POTENTIAL TREATMENT TECHNOLOGIES FOR THE NFSS K-65 RESIDUES

Based on the 1996 treatment technology screening performed by DOE, information gathered from the FEMP treatment technology evaluations, and a literature review of recent treatment technology developments four technologies are recommended for further evaluation for the NFSS K-65 residues: 1) reclamation (resource/recovery), 2) stabilization, 3) encapsulation, and 4)\_photodeactivation (transmutation using gamma rays) The first three technologies are conventional technologies that have been used for hazardous and radioactive waste treatment. The fourth technology, photodeactivation, is an extremely innovative technology that has not been used in the past to treat radioactive waste. The reclamation, stabilization, and encapsulation technologies, potential vendors, and the basis for their selection are described in Sections 2.4 through 2.6. The photodeactivition technology is described in Appendix A. A summary of the benefits and potential drawbacks offered by these technologies is presented in

NFSS – USACE Tech Memo: Treatability Studies for NFSS November 2002 Table 2.1. Each of these technologies could be different components of various alternatives under consideration for NFSS.

Abbreviated work plans for treatability studies to evaluate reclamation, stabilization, and encapsulation are presented in Section 4. Appendix A contains a brief description of the transmutation technology.

To obtain implementation details for the abbreviated work plans, three vendors (listed below) were contacted and are discussed in this document however, other vendors may have similar capabilities:

- 1) Reclamation International Uranium Corporation (IUC);
- 2) Stabilization Perma-Fix Environmental Services (using Perma-Fix 1®)
- 3) Encapsulation Eurotech (using EKOR<sup>TM</sup>).

Table 2.2 Technology Benefits and Possible Drawbacks

	Table 2.2 Technology Benefits and Fossible Drawbacks				
Technology	Benefits	Possible drawbacks			
Reclamation at IUC	May provide cost avoidance opportunities (recycling of transportation containers, disposal of secondary waste in tailings pond)	<ul> <li>Contract issues and legal liabilities may govern or override any technical considerations</li> <li>NFSS stakeholders may not be receptive (FEMP stakeholders rejected this as a legitimate treatment alternative)</li> <li>The market for the recovered uranium at the time of reclamation can not be determined</li> <li>Initial IUC evaluations suggest that a 40:1 (residues:lower activity waste) blend ratio would be required prior to reclamation</li> </ul>			
Solidification/ Stabilization (using Perma Fix or similar stabilization technology)	<ul> <li>Conventional and straightforward technology</li> <li>Proven technology used extensively in full-scale applications</li> </ul>	<ul> <li>May not be adequate space at the NFSS site for set-up of the mobile treatment system</li> <li>Treatment will increase waste transportation and disposal volume</li> <li>Stakeholders may object to on-site treatment</li> <li>May require management and disposal of secondary waste based on residue removal method used</li> </ul>			
Encapsulation (using EKOR <sup>TM</sup> or similar process)	<ul> <li>May provide higher waste loadings than conventional stabilization/solidification agents</li> <li>May provide shielding benefits</li> <li>Applicable to wastes in both dry and slurry form.</li> </ul>	<ul> <li>Full-scale use of technology as an encapsulation agent for radioactive waste is limited</li> <li>May not be adequate space at the NFSS site for set-up of the mobile treatment system</li> <li>Treatment will increase waste transportation and disposal volume</li> <li>Stakeholders may object to on-site treatment</li> <li>May require management and disposal of secondary waste based on residue removal method used</li> <li>May be expensive</li> </ul>			
Photodeactivation	May significantly reduce the volume of waste requiring disposal	Unproven technology for radioactive waste treatment     Application in a full-scale operations may require lengthy     development time			

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### 2.4 RECLAMATION

### 2.4.1 **Technology Description**

One of the potential technologies recommended for further evaluation is reclamation of the uranium and other valuable metals that may exist in the residues. IUC's White Mesa Mill located outside Blanding, Utah appears to offer the greatest potential for providing resource recovery of the NFSS residue. IUC is an NRC licensed facility that has the capability to process natural uranium-bearing ores and alternative feed materials to extract the uranium, vanadium and other valuable resources that may exist in the material.

It should be noted that radium and thorium recovery was discussed with IUC. Due to the complexities associated with extracting these constituents and lack of economic incentive, it was not considered feasible to recover radium and thorium from the residues. In addition, historical documents (DOE 1984) indicate that the ability to decontaminate recovered metals to the degree necessary for subsequent marketing might be difficult.

Conventional ore uranium extraction processing involves physically grinding the ore (with the addition of water) to form a slurry containing approximately 50% solids followed by a preleaching and leaching process to dissolve the uranium from the slurry. The pre-leaching of the slurry is performed using a strong acid solution followed by a pre-leach thickening using flocculants to separate the solids from the uranium rich solution. The underflow solids are transferred into the second stage leach circuit where acid, heat, and oxidant are added to obtain additional uranium recoveries. The uranium rich solutions from the pre-leach and leach steps are sent to the solvent extraction circuit where the uranium is selectively removed from the acidic water solution with an organic solvent. The uranium is concentrated in the solvent organic phase and the aqueous solution (i.e., raffinate) is barren in uranium. The raffinate is pumped to the tailings ponds and the organic solution is pumped to the stripping circuit where the uranium is stripped from the organic and again concentrated. The loaded high-grade strip solution is pumped to the precipitation circuit where the addition of ammonia, air, and heat causes the uranium to become insoluble and precipitate out of solution as "yellow cake" (U<sub>3</sub>O<sub>8</sub>). Figure 2-1 presents a simplified process flow diagram (PFD) of the extraction and recovery process.

The processing of alternate feed materials (such as the K-65 residues) is very similar to the conventional uranium extraction process described above with the minor modifications described here. If required, the alternate feed material is passed through a 30-foot by 6-foot diameter trommel screen prior to introduction into the leach circuit. Water is added to wash the feed material, break up large lumps, and remove debris and organic material prior to introduction of the feed to the mill. The leaching conditions will be dependent on the characteristics of the feed material. For alternate feeds the leach conditions range from 1 to 24 hours retention time and operating temperatures range from ambient to 100 degrees Celsius. For alternate feed material processing the uranium is extracted from the clarified pregnant liquor through an ion-exchange process or a solvent extraction process or a combination of the two.

# 2.4.2 Justification for Selection

Reclamation was recommended for further evaluation as the technology provides an alternative to more traditional treatment technologies and may offer cost avoidance opportunities (such as recycling of transportation containers, avoided disposal costs, etc.).

IUC was selected as the potential vendor as they have operated a successful uranium processing facility for over 20 years, they have processed Formerly Utilized Sites Remedial Action Program (FUSRAP) materials containing similar uranium concentrations as the K-65, L-30, and L-50 residues, they have processed FUSRAP materials containing Ra<sup>226</sup> concentrations similar to the L-50 residues, and they have licenses that could be amended to allow the receipt and processing of the NFSS residues as an alternate feed material.

Natural uranium ores with higher activity levels have been successfully processed in conventional uranium mills. In Canadian uranium milling facilities, conventional mined ores containing 4% uranium have been successfully processed while maintaining occupational doses and radioactive releases to the environment at acceptable levels. Such ores result in external gamma radiation levels calculated at approximately 15 mrem/hr to tissue in contact with drums of these ores, and up to approximately 1.5 mrem/hr to tissue at a distance of 1-m from a drum. For these ores, Ra<sup>226</sup> and Th<sup>230</sup> concentrations are each approximately 13,200 pCi/g. This experience demonstrates that processing of such ore grades is feasible using conventional milling technology; however, IUC has not had the opportunity to process these higher concentration materials.

IUC possesses an NRC Materials License that allows the licensee to receive, acquire, possess, and transfer byproduct, source, and special nuclear material and to process such material in its White Mesa uranium milling facility. To receive and process the K-65 residues IUC would need to obtain a material-specific license amendment, in accordance with 10 CFR 40, that would allow IUC to receive "Alternate Feed Material" other than natural uranium ore, for processing in the White Mesa Mill. License amendment requests are approved by NRC on a case-by-case basis. During the past ten years, IUC has requested fourteen, and received thirteen license amendments to process alternate feed materials (the fourteenth is currently under consideration by the NRC) (IUC 2002).

White Mesa Mill is licensed to process up to 2,000 tons per day (tpd) of uranium bearing ore. The Mill has eight high capacity thickeners, which are capable of being configured into groups or series of parallel stages. Three separate solvent extraction (liquid ion exchange) circuits are capable of handling aqueous flows up to 800 gallons per minute. Final products can be dewatered, dried, or calcined at temperatures up to 650 degrees centigrade. The Mill operates on a "processing campaign" basis (e.g., material is received and accumulated over a period of time until a sufficient amount to process is obtained on-site). Then a processing campaign is undertaken until the stored material has been processed. Maintenance and upgrades of the Mill are conducted during the downtime between processing campaigns.

The Mill has approximately 20 acres of storage space within the Mill's Restricted Area available for bulk or containerized materials. The Mill can accommodate high moisture content materials and most forms of debris consequential to excavation activities (e.g., concrete, asphalt, etc). Waste streams from the process are pumped to the Tailings Management System that currently consists of four below grade tailings cells lined with synthetic liners and leak detection systems. The tailings system is a zero-discharge system separated by nearly 1,200 feet

of low-permeability rock from the regional aquifer. Based on over 20-years of monitoring, the Mill's tailing system has performed in accordance with applicable regulatory requirements.

Over the last five years IUC has received over 283,000 tons of FUSRAP materials, processing more than 47,000 tons of this material in 1999. Additional processing of FUSRAP material is scheduled to start in June 2002.

# **Application in Full-Scale Operations**

IUC's White Mesa Mill would be the location for processing the bulk NFSS residues during the full-scale operations. The treatment operation is described below.

Residues would be excavated from the NFSS containment cells and downblended on-site at NFSS to allow the residues to be shipped in accordance with DOT requirements to White Mesa Mill. The NFSS residues would be transported to White Mesa Mill either by truck or intermodal (i.e., rail plus truck) because there is no rail spur to the facility. The nearest rail offloading site is Green River, Utah, 132 miles to the northwest. Other rail sites, used for transportation of alternative feed materials are Cisco, Utah, 140 miles to the north and East Carbon, Utah, 185 miles to the northeast. To date, over 283,000 tons of FUSRAP material have been transported to the White Mesa Mill through the Cisco or East Carbon railheads.

Results of recent treatability studies at the Fernald Environmental Management Project (FEMP) indicate that the waste must be downblended to a waste loading of 17 % to cost effectively meet DOT requirements. It is assumed that the high-activity NFSS residues (i.e., K-65, L-30, L-50) residues would be blended with lower-activity materials (R-10, NFSS remaining waste) on site before shipment to IUC. Once at the White Mesa Mill the NFSS wastes is likely to require further downblending to produce an alternate feed material with target activity levels comparable to conventional mined ores (activity levels of approximately 13,200 pCi/g Ra<sup>226</sup> and Th<sup>230</sup>). Preliminary evaluations of the characteristics of the K-65 residues suggest that a 40:1 blend ratio may be required (40 parts low-activity material to 1 part K-65 residues) to maintain occupational doses and radioactive releases to the environment at acceptable levels.

Incoming waste shipments would be weighed and the waste would be transported to the Mill's pulp (wet) storage tanks. When a sufficient quantity of waste is on-site, the blended alternate feed would then be processed through the leach circuit. After processing, IUC would package the yellow-cake in 55-gallon drums and ship it to the converter facility (i.e., Allied Signal facility in Metropolis, Illinois) where it would be converted to UF6 for utilities or other customers. The USACE would not physically own the U<sub>3</sub>O<sub>8</sub> after processing. IUC has implemented the practice of sending the yellow-cake off-site as soon as practical based on the processing rate and scheduling for shipment. Currently, it is expected that the material would be on-site from 1 to 2 weeks at most prior to being sent to the converter facility, with the length of time dependent on market demand and utility customer contracts. While at IUC, the yellowcake would be stored within locked fencing and under 24 hour security.

Byproducts from the processing operations would be sent to the Mill tailings cell.

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# 2.5 SOLIDIFICATION/STABILIZATION

# 2.5.1 Technology Description

The second potential technologies recommended for further evaluation is solidification/stabilization. Stabilization changes the chemical properties of the treated material through chemical reactions. Solidification incorporates the contaminants into a solid matrix.

The solidification/stabilization treatment process proposed for further evaluation is the Perma Fix 1® process. The Perma-Fix 1® solidification/stabilization process begins by adding water to the material to create a slurry. Then, a fixating agent of sulfide material is added to the waste to chemically bind the RCRA hazardous constituents as well as radioactive metals such as uranium and thorium. The chemical reactions precipitate the leachable metals in the material into a highly insoluble form. Additional additives (absorbents/sealants) can be used to reduce emission of radon from the material. The final step dries the slurry through absorption and/or a chemical reaction and converts the waste to a solid, monolithic or loose form by microencapsulation in a cement matrix. This step provides an additional barrier to leaching (Perma-Fix 2002).

## 2.5.2 Justification for Selection

Solidification/stabilization was selected for further evaluation as the technology has the potential to treat the K-65 residues to meet DOE disposal facility WAC should a commercial disposal facility capable of accepting 11(e)(2) wastes become unavailable. The treatment process can also provide shielding against radioactive emissions from the material, thereby decreasing the radiation risk to the material handlers and the public. The Perma-Fix 1® process targets the RCRA hazardous characteristics and radioactive metals that are found in the K-65 residues and it can decrease specific activity by blending the material with cement.

Perma-Fix Environmental Services was selected as a potential vendor because of both experience and capability in that they have treated similar residues in the form of sump cake at the FEMP and they have the ability to treat the material onsite, which may result in a reduction to the overall remedial action costs at NFSS. However, other vendors can provide similar services.

Perma-Fix Environmental Services, Inc. provides hazardous and mixed wastes treatment to commercial and government customers. The company operates three RCRA-permitted, radioactive licensed facilities designed for the treatment of mixed waste and also have the capability for on-site remediation. Perma-Fix Environmental Services could perform treatability studies at Perma-Fix of Florida (PFF). PFF has a Radioactive Material License (number 2598-1) from the State of Florida, and a hazardous waste permit for the receipt and treatment of RCRA characteristic hazardous waste and certain F-listed and U-listed hazardous wastes.

Solidification/stabilization has been selected as the preferred treatment alternative for the K-65 residues at the FEMP site. Much information is available from FEMP on solidification/stabilization of the K-65 residues; however, there are many site-specific process variables that will determine its effectiveness and cost for the NFSS waste. (For example, the FEMP Silos 1 and 2 contain K-65 residues and BentoGrout clay. BentoGrout clay is not present in the NFSS K-65 residues. In addition, the removal method selected for the NFSS

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residues will impact the solidification/stabilization treatment process unit operations and the recipe.)

# 2.5.3 Application in Full-Scale Operations

PFF has the ability to design and construct waste treatment systems for on-site treatment at sites such as NFSS (Perma-Fix 2002). Residues would be removed from the NFSS containment cells and treated by Perma-Fix on-site at NFSS. The method of residue removal (i.e., slurry vs. some other mechanical means) will determine the exact equipment configuration. For purposes of discussion, the treatment system would convey the waste into residue storage bins by a screw conveyor. Waste from the bins would be fed through a delumper and then into a batcher. A computer system would control the amount of waste, Portland cement, and liquid reagent that is then combined in a mixer. If a monolithic final form is desired, the slurry would then be poured into containers and cured for approximately 24 hours (Perma-Fix 2002). Cured waste forms would be tested to ensure they meet disposal facility WAC then shipped off-site for disposal. When waste processing operations are complete the treatment system would be decontaminated, disassembled, and returned to the Perma-Fix facility or disposed.

In order for the results of a treatability study to be useful for full-scale application, the physical parameters (i.e., moisture content, residue/onsite soil mixture) of the waste stream must be known or accurately estimated. Estimation of the physical parameters of the residues would be difficult at this time as the method of residue retrieval (i.e., direct excavation, remote retrieval in slurry form, etc.) from the WCS is unknown.

# 2.6 ENCAPSULATION/CONTAINMENT

# 2.6.1 Technology Description

The third technology proposed for further evaluation is encapsulation using the EKOR<sup>TM</sup> process. This product was identified by USACE Buffalo district personnel and was considered for potential applicability at NFSS. This relatively new product is marketed for use in containment and encapsulation, stabilization, and as a shielding agent. This product has the potential of being a substitute for conventional stabilization materials, such as cement/grout, with an added potential of being used as a spray or preformed sheet application for increasing radiological shielding.

EKOR<sup>TM</sup> is a silicone block copolymer exhibiting high resistance to radiation without becoming radioactive after exposure and shows little degradation due to aging or chemical exposure. It has very low permeability, no measurable leachability, and contains no toxic components. It was created by a team of nuclear scientists from the I.V. Kurchatov Research Center in Russia and the EuroAsian Physical Society to address the radioactive concerns from the 1986 accident of Reactor 4 at Chernobyl, Ukraine.

# 2.6.2 Justification for Selection

Encapsulation using the EKOR<sup>TM</sup> process was selected for further evaluation as the technology has the potential to treat the K-65 residues to meet disposal facility WAC and based on vendor information may offer much higher waste loadings (up to 70%) than conventional

NFSS – USACE Tech Memo: Treatability Studies for NFSS November 2002 solidification/stabilization products. Materials such as boron or tungsten can be incorporated into the mixture to increase the shielding properties. This product also has the potential of being used as a possible spray or preformed sheet for increased radiological shielding. US-based Eurotech, Ltd. owns the rights to EKOR<sup>TM</sup>.

The product has demonstrated ability to encapsulate material of high radiological properties. These demonstrations include:

- Encapsulation or "cocooning" of the most critical radioactive fuel containing masses resulting from the Chernobyl disaster. Coating the mass successfully prevented radioactive material from dusting or seeping into the environment.
- Encapsulation of wastes in both dry and slurry form.
- Encapsulation of radioactive debris at the Savannah River Site (SRS), a DOE facility near Aiken, South Carolina, managed by Westinghouse Savannah River Company (WSRC).

# 2.6.3 Application in Full Scale Operations

Limited information was available to evaluate how the EKOR<sup>TM</sup> process would implemented in a full-scale operation however, it is expected that the implementation would be similar to that described in Section 2.5.3 for the Perma-Fix  $1^{\text{@}}$  process.

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### 3.0 STUDY REQUIREMENTS

### 3.1 SAMPLE COLLECTION

A sample of the K-65 residue currently within the WCS is not available, and retrieval of a sample from the WCS is not currently contemplated. The use of a surrogate in performing the treatability studies was contemplated for this project. However, representatives at FEMP (SAIC 2002) indicated that complex interaction between the K-65 residue constituents were not adequately represented in the surrogate mixture. This was discovered after the surrogate tests were performed, and the results used in the design of a full-scale vitrification unit. Since the results were not comparable to actual K-65 residue behavior, the FEMP vitrification project encountered many scope changes and redesigns. Therefore, FEMP has decided to use actual K-65 residue in determining treatment formulas for stabilization/solidification, which is now the preferred alternative for K-65 remediation.

The K-65 residue present within Silo 1 at the FEMP is believed to be similar to the residue in the WCS at NFSS however, existing characterization for the two materials is limited. A comparison of available characterization data is shown in Table 3-1.

Table 3-1. Comparison of the Radiological and Chemical Constituents of the NFSS K-65 Residues with the FEMP Silo 1 Material

G 11	NEGG	G 45	555.45	G 4D
Constituent	NFSS	Source of Data	FEMP	Source of Data
Radium (average)	520,000 pCi/g	DOE 1986	391,000 pCi/g	DOE 1994
Radium (range)	180-217 ug/kg	Dettorre 1981	200-360 ug/kg	Dettorree 1981
U-238 range	470-650 pCi/g	DOE 1986	387-920 pCi/g	DOE 1984
U range	500-30,000 mg/kg	DOE 1986	600-3,200	Dettorree 1981
Arsenic (average)	5	DOE 1986	22	DOE 1993
Barium	30,000	Bechtel 1984	50,000	Litz 1984
Cadmium (average)	2.5	DOE 1986	2	DOE 1993
Chromium (average)	100	DOE 1986	42	DOE 1993
Cobalt	2,000	Bechtel 1984	1,600 - 2,000	Litz 1984
Copper	1,480	Bechtel 1984	500 - 800	Litz 1984
Gold	9	Bechtel 1984	65 - 78	Litz 1984
Lead	56,000	DOE 1986	48-70,000	Litz 1984
Mercury	0.5	DOE 1986	0.6	Litz 1984
Nickel	3,700	Bechtel 1984	3,500 - 3,700	DOE 1993
Palladium	27	Bechtel 1984	13 – 18	Litz 1984
Platinum	0.6	Bechtel 1984	0.9 - 1.4	Litz 1984
Silver (average)	1.5	Bechtel 1984	18	Litz 1984

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Table 3.2. Comparison of Physical Properties of K-65 Residues at NFSS and FEMP

	Grain Size Distributions						
NI	FSS		FEMP				
Size	%	Size	1-1	1-2	1-3	1-4	Average %
>840 um	4.1	>850	6.8	8.4	4.9	4.5	6.2
840-240	9.5	850-250	1.6	10.4	13.1	14.4	9.9
240-150	1.8	250-106	11.5	9	34.3	22.4	19.3
150-74	5.2	106-75	7.4	0.7	5.4	4.2	4.4
74-44	4	75-47*	23.4	9.4	15.4	13.8	15.5
44-37	2.3	47-34**	3	3.6	4.8	3.1	3.6
<37	73.1	<34***	46.3	58.5	23.7	37.6	41.5

FEMP data taken from an IT certificate of analysis dated March 22, 1990.

All are Silo 1 samples with the fourth being a composite number of three other samples (not the previous three).

Based on the limited characterization data and historical process knowledge it is believed that using a sample of the FEMP Silo 1 material would meet the needs of the NFSS treatment technology evaluations. At the beginning of the preparation of this technical memorandum in February 2002, initial discussions with FEMP representatives indicated there might be sufficient K-65 residue in storage at FEMP that may be available to USACE for use in treatability studies. Upon further discussions with FEMP representatives in late May 2002, it was concluded that a sample is not available for use by the USACE. In addition, obtaining a sample for USACE use would require a formal request from the USACE to DOE and agreements regarding which agency is liable for management of the sample and disposal of the residues from the treatability studies would need to be negotiated. Reaching such agreements between the USACE and DOE will likely be complex and time consuming.

Obtaining additional waste samples from Silo 1 also does not currently appear feasible. Health and safety and logistical considerations appear to make opening the Silo for obtaining a small amount of sample for USACE treatability studies unrealistic. Because of the significant chance of radon gas in the headspace of the silo, and the significant health and safety considerations required to deal with this issue, extensive planning would be required to obtain a sample. Extensive coordination would be required with DOE, and assistance for a project that provides no benefit for DOE is anticipated to be difficult to obtain. A work-plan would be required as well as a Radioactive Work Permit submitted and issued, with extensive review and approval from DOE and FEMP personnel. Significant labor costs (estimated at over \$250,000) are anticipated. Costs would include compensation for personnel exposure related to sample collection that may prohibit future radiological exposure during the calendar year for the sample crew.

Within the next two to three years, it is anticipated that Silo 1 will be opened to implement the remedial action for the residues. Residue material may then be available for USACE use in the treatability studies.

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Each sample had a different hydrometer analysis ( $<75 \mu m$ ). Values for the lower end range from 51-42  $\mu m$ .

<sup>\*\*</sup> Hydrometer analyses were for 34-47, 31-42, 36-51, and 35-49 μm.

<sup>\*\*\*</sup> This value was 34, 31, 36, and 35  $\mu$ m for samples 1, 2, 3, 4, respectively.

# 3.2 SAMPLE SHIPMENT REQUIREMENTS

Preliminary evaluations of shipping requirements indicate that treatability study samples of the K-65 residues could be transported as a "Limited Quantity" shipment via Federal Express (air or ground) if properly packaged such that dose readings on the surface of the outer package do not exceed 0.5 millirem/hour (mrem/hr). Limited quantity shipment designation is based on the radionuclides present in the material (measured as total curies of the specific radionuclide) compared to DOT standards under 49 CFR §§173.421 and 175. Specific marking and labeling requirements enumerated under these regulations must be addressed at the time of shipment.

In order to meet the limited quantity contact surface dose rate the sample must be packaged in an inner container with sufficient shielding to attenuate any gamma radiation to this level. The packaging/shielding must not shift during transport resulting in an increase in surface dose rate over the specified limit.

If sufficient shielding cannot be provided to meet this dose rate, the sample will then have to be shipped as a low specific activity (LSA II) shipment. This classification requires that the sample be shipped in a Type A container and additional shipping documentation, labeling, and notification must be provided.

Cost for shipment of the K-65 residue material, assuming the above referenced designations apply, is not expected to exceed \$500. Final determination of approved packaging and other shipping requirements would be made by a qualified shipper at the time of shipment of the FEMP Silo 1 material.

# 3.3 DISPOSAL FACILITY SELECTION

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Two of the treatment technologies (stabilization/solidification using Perma-Fix and encapsulation/shielding using EKOR<sup>TM</sup>) generate a treated matrix that will require off-site disposal of the K-65 NFSS residues. For the purpose of this treatment technology evaluation target disposal facilities for the treated residues needed to be selected. to determine disposal facility WAC.

While disposal of the treated K-65 residue can be physically accomplished in a safe manner at several facilities, the ability to dispose of the material in a manner that satisfies state/local statutes and/or DOE policy/requirements is very limited. Three disposal facilities, Hanford Storage Site, NTS, and Envirocare of Utah (Envirocare), currently possess the necessary licenses and permits to accept the treated K-65 residue. However, NTS is currently only permitted to dispose of low-level radioactive waste from on-site locations and other DOE sites. (A permit application to accept mixed low-level waste from other DOE facilities was submitted to the Nevada Division of Environmental Protection on December 22, 2000). The Hanford Storage Site does not accept mixed waste from other DOE sites. (The approval to accept mixed waste from other DOE sites is pending completion of an environmental impact statement).

Because the completion of the NFSS remedial action has been transferred from the DOE to the USACE, disposal of the treated residue at a DOE facility was considered a secondary option, and focus for this evaluation was placed on a commercially available disposal facility. Therefore, Envirocare was selected as the most viable facility for commercial disposal of the treatability study wastes. Envirocare has an Nuclear Regulatory Commission (NRC) License

that allows the acceptance of 11(e)(2) material. Material classified as 11(e)(2) byproduct material would be disposed of in a waste cell specifically designated for such material. The acceptance limits for radioactivity are different for 11(e)(2) material than for radioactive or RCRA mixed waste, and depend on amounts of radioactivity per truckload. Any truckload or railcar containing waste with an average concentration above 4,000 pCi/g of any radionuclide in the <sup>226</sup>Ra series cannot be accepted. Envirocare has requested a license amendment to allow the 11e(2) cell to accept up to 100,000 pCi/g of Ra-226. If this amendment is approved by the NRC, the K-65 residue may meet the disposal facility WAC in their current form without treatment. Because Envirocare is currently the most viable option available for the disposal of the K-65 residues, the abbreviated treatability study work plans for Perma-Fix and EKOR<sup>TM</sup> were developed to satisfy the Envirocare WAC.

Disposal facility needs for transmutation will be evaluated in an addendum to this report.

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# 4.0 ABBREVIATED TREATABILITY STUDY WORK PLANS

This section contains abbreviated treatability study work plans for three treatment technologies identified for further evaluation (reclamation, stabilization, encapsulation). Information regarding the fourth technology identified for further evaluation, photodeactivation, is contained in Appendix A. Information presented in this section was obtained from the technology vendors or developed based on reviews of historical information regarding the characteristics of the NFSS waste and the results of the treatability studies performed by FEMP on the K-65 residues. During the evaluation, SAIC interfaced with the treatment vendors to establish treatability objectives and tests to be performed. Historical analytical data was provided to each of the vendors (IUC, Perma Fix, and Eurotech) for review. SAIC requested that each vendor provide a work plan of sufficient detail that described how the vendor would accomplish the treatability study. The level of detail varies among the three work plans presented in this section based on the amount of information supplied by the vendor. To the extent possible information is presented to address the elements recommended by EPA for a treatability study work plan (EPA 1992). Information presented in other sections of this document (such as technology descriptions) is not repeated here.

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Alternate Feed Stockpile Trommel Screen Pre-Leaching Thickening Leaching **Uranium Bearing Solutions**  $U_3O_8$ Counter Extraction Current Washing Barren Raffinate Solids  $U_3O_8$ Stripping **Tailings** Yellow Cake Precipitation & Thickening **IUC White Mesa Uranium Mill** Drying & Blanding, Utah Packaging **Uranium Product** to Shipment

Figure 4.1 International Uranium (USA) Corporation Alternate Feed Process Flowsheet

# 4.1 WORK PLAN FOR RECLAMATION AT INTERNATIONAL URANIUM CORPORATION

# 4.1.1 Treatability Study Location

Treatability testing is performed at IUC's White Mesa Mill located approximately six miles outside Blanding, Utah. The White Mesa Mill has metallurgical and analytical laboratory facilities and certified personnel capable of performing treatability studies. The laboratory is covered under IUC's NRC Materials License. The laboratory has the capability to perform size reduction, blending, leaching, metals assaying, and volumetric analysis of the leach product and leaching residues. IUC's contract laboratories would perform additional analyses required for the treatability study.

# 4.1.2 Test Objectives

The primary objective of a reclamation treatability study would be to determine

- 1) the amount of downblending required for the residues to meet DOT and IUC requirements for waste shipments offsite and for processing
- 2) the feasibility of reclaiming uranium from the residues,
- 3) the uranium removal efficiency,
- 4) the quantity and characteristics of secondary waste generated,
- 5) the health and safety procedures required to handle, process, and dispose of the process residues, and
- 6) the estimated costs of the reclamation for full scale production (including downblending, transportation, reclamation, storage and disposal costs).

A secondary objective could be the evaluation of the feasibility of reclaiming other valuable metals from the residues.

# 4.1.3 Experimental Design

The treatability study would be structured to produce a blended sample of K-65 material and alternate feed material that contains target activity levels comparable to materials previously processed at conventional uranium mills and to assess parameters that may affect the operability, safety, and cost of processing the NFSS residues. The following basic steps would be performed during the study:

- Blend a sample of K-65 material with lower-activity alternate feed material to produce a sample with the proposed activity levels. The blended sample would need to have the characteristics of onsite NFSS soils or other downblending materials that would be used at NFSS.
- Subject the blended sample to various bench-scale leaching processes to determine the appropriate leaching/digestion chemistry and leaching efficiency;
- Determine the appropriate extraction circuit configuration and extraction efficiency;
- Assess product quality and overall uranium recovery at optimum operating conditions;

• Determine the appropriate physical handling steps for transfer, stockpiling, and introduction of the material to the leach circuit, and determine environmental and worker safety impacts from material and waste handling activities; and

The analytical data generated from the treatability study would include:

- Chemical, physical, and radioactive characteristics of both the K-65 sample, the pre-mix alternate feed material to be blended with the K-65 sample, and the blended sample prior to the treatability study;
- Weight percent of uranium and any other metal assays of value in the K-65 sample, the pre-mix alternate feed material, and the blended sample;
- Uranium content, activity levels of key radionuclides, and other metals content in the waste streams from the treatability study; and
- Uranium content, activity levels of key radionuclide, and other metals content in the product from the treatability study.

The treatability study results would be used by IUC to evaluate unit reclamation processes (leaching, solvent extraction/ion exchange, precipitation, etc.), establish processing conditions, and determine the cost of full scale processing of the NFSS residues. The results would also be used to evaluate whether the process byproducts could be disposed in IUC's tailing cells.

One kilogram of sample is needed by IUC to perform an effective treatability study on the K-65 residue.

# 4.1.4 Residuals Management

After completion of the treatability study any used treatability study sample and residues generated during the study would be disposed of in IUC's tailings disposal facility.

# 4.2 WORK PLAN FOR STABILIZATION/SOLIDIFICATION USING PERMA-FIX 1®

# 4.2.1 Treatability Study Location

Treatability testing is performed at PFF located in Gainesville, Florida. PFF is a full-scale treatment facility with an analytical laboratory. The facility can accept radioactive, hazardous, and mixed wastes in the form of liquid, solid, sludge, debris, or soil.

# 4.2.2 Test Objectives

The primary objective of a stabilization treatability study would be to determine 1) the feasibility of achieving disposal facility waste acceptance criteria, 2) the maximum waste loading for the solidification/stabilization process, 3) the waste volume increase, 4) shipping requirements for the treated waste forms, 5) the quantity and characteristics of the secondary waste generated, 6) the health and safety procedures required to perform the treatment operations and dispose of the

secondary waste, and 7) the estimated costs for the full scale solidification/stabilization process (including treatment, transportation, and disposal costs).

# 4.2.3 Experimental Design

The treatability study sample should be as close as possible to the same physical form as the material at NFSS would be after removal. As the removal method for the K-65 residues has not been determined PFF assumed a 70% moisture content value as the target to simulate the probable condition of the bulk residues at NFSS. (Note that the 70% moisture content value assumes that the residues would be hydraulically mined from the current subsurface location within the WCS at NFSS.)

The following basis steps would be performed during the study:

- The treatability sample would be characterized.
- The sample would be subjected to a two-step stabilization and solidification process. The first step would precipitate leachable metals from the slurry in a highly insoluble form. In the second step the slurry would be dried through adsorption and/or chemical reaction.
- Several batch tests would be run to optimize treatment parameters.
- Once the best treatment protocol is established three aliquots of waste will be treated individually to produce treated samples for third party analysis.

The treatability study results would be used by PFF to establish processing conditions, determine the cost for full scale processing of the NFSS residues, and determine shipping requirements for the treated waste. The results would also be used to evaluate the disposal of secondary waste generated by the solidification/stabilization process.

The amount of sample material needed depends on parameters to be analyzed and the level of study needed. For the proposed bench scale study approximately five gallons (24 kg) would be sufficient.

# 4.2.4 Residuals Management

PFF would dispose of analytical laboratory wastes, extracts, and secondary wastes (e.g. personal protective equipment, disposable lab ware, empty containers, etc.) generated by PFF and the third party laboratory during the study. Excess sample material not used for analysis by PFF's contract laboratory would be returned to PFF once the analytical results are reviewed and accepted by PFF. (Perma-Fix 2002)

PFF would return all treated and untreated sample materials to the generator (agreements would be needed to designate generator and liability status). No additional hazardous waste codes would be added to the sample through the treatment process. It was assumed that the treatability study residuals would be sent to Envirocare for final disposal.

# 4.3 ENCAPSULATION AND SHIELDING USING EKOR<sup>TM</sup>

# 4.3.1 Treatability Study Location

The treatability study would be conducted at Nuclear Fuel Services (NFS), located in Erwin, Tennessee, as a subcontractor to Eurotech, Ltd. NFS specializes in nuclear fuel fabrication and has been in operation since 1957. NFS has conducted several treatability studies for various customers including the DOE and the Department of Defense. As a teaming partner with FEMP for remediation of the site, NFS performed a variety of treatability studies for FEMP mixed waste streams. Many of these streams contained heavy metals that were treated to comply with LDRs.

The NFS Applied Technology Laboratories include a 3,500-ft<sup>2</sup> bench/pilot scale test area and a 1,500 ft<sup>2</sup> analytical laboratory. The analytical laboratory is used to the support treatability studies and has the capability to perform radiochemistry and chemistry.

NFS has a Special Nuclear Materials License SNM-124 for operations using enriched uranium, from nuclear fuel manufacturing to high-enriched uranium processing and blend-down. Their Development Laboratories also possess a radioactive materials (source and by-product) license issued by the State of Tennessee, No. R86009J97. These laboratories operate in accordance with the licenses designated above, as well as under the treatability exemption regulations provided by 40 CFR and the Tennessee Hazardous Waste Regulations. Under these regulations, NFS may accept up to 10,000 kg of media contaminated with RCRA hazardous waste for treatability studies (Eurotech 2002b).

# 4.3.2 Test Objectives

The primary objective of an encapsulation treatability study would be to determine 1) the feasibility of achieving disposal facility waste acceptance criteria, 2) the maximum waste loading for the EKOR<sup>TM</sup> process using moist residues, 3) maximum waste loading for the EKOR<sup>TM</sup> process using various slurry moisture contents, 4) the waste volume increase, 5) shipping requirements for the treated waste forms, 6) the quantity and characteristics of the secondary waste generated, 7) the health and safety procedures required to perform the treatment operations and dispose of the secondary waste, and 8) the estimated costs for the full scale EKOR<sup>TM</sup> process (including treatment, transportation, and disposal costs). A secondary objective that will be evaluated is whether EKOR<sup>TM</sup> can be used as an effective shielding material either mixed with the residue or applied to waste storage and transport containers.

# 4.3.3 Experimental Design

The following basic steps would be performed during the treatability study:

- Waste will be homogenized to the maximum extent practicable.
- Samples would be collected from the K-65 residues and initially characterized for metal TCLP components. Samples would then be collected and forwarded to a contract laboratory for certified analyses of TCLP components and radiological analysis. The purpose of these tests would be to establish appropriate reagent recipes for each specimen and to provide for an initial indicator of waste loading.

- The product would be mixed with the wet waste with up to 70% waste loading, using a stand impeller mixer or larger drum mixing equipment. NFS would evaluate different waste loadings and perform these tests in duplicate.
- The resultant mix would then be cured for 48 to 72 hours.
- TCLP analysis would be performed on the encapsulated material from these tests. The TCLP leach fluid would be assayed for key radionuclides such as <sup>210</sup>Pb and <sup>226</sup>Ra. Select samples of the treated waste would be sent to a certified laboratory for analysis of TCLP constituents and key radionuclides.

NFS recommends that a minimum of 15 kg and not more than 25 kg of K-65 residue be provided for the treatability study testing. Eurotech, Ltd. would provide the polymers and catalysts for the treatability testing. It should be noted that for obtaining shielding properties, any gamma-emitting source may be used for the evaluation.

NFS would also work with Eurotech, Ltd to identify other mechanisms for applying the product to the K-65 residue (e.g., with additives for shielding or potentially as a sealant). This testing could involve coating containers containing K-65 residue to determine the corresponding reduction in the radiological field. The product would also be applied in sheets using varying amounts of additives (such as iron and tungsten) to determine the field reduction with the optimal product-to-additive ratio.

A valuable feature of the EKOR<sup>TM</sup> product is its potential ability to reduce the radiological field emitted from the treated or untreated K-65 residue. The study would evaluate the reduction in field once the residue is absorbed into the matrix as well as when used as a coating. The treatability study would determine the most effective and efficient method of reducing radiological fields in containers as well as the possibility of having containers walled with product (pure or with additive) sheets.

# 4.3.4 Residuals Management

All treated and untreated residues would be packaged and returned to the generator no later than one year after receipt of the sample for testing or 90 days after completion of the treatability study, whichever occurs first. NFS maintains a qualified transportation subcontractor and would arrange for transportation of the material back to the generator.

# 5.0 CONCLUSIONS

For the purpose of this technical memorandum the evaluation of potential treatment technologies and treatability studies for the NFSS focused on treatment of the K-65 residues. These residues represent the greatest technical challenge at the site, contain the highest activities, and were the subject of NAS recommendations.

Based on a review of the previous technology screening performed by DOE, information gathered from the FEMP treatment technology evaluations and recent literature, four technologies are recommended for further evaluation for the NFSS K-65 residues: 1) reclamation (resource/recovery), 2) stabilization, 3) encapsulation, and 4) photodeactivation (transmutation using gamma rays). The first three technologies are conventional technologies that have been used for hazardous and radioactive waste treatment. The fourth technology, photodeactivation, is an innovative technology that may have application for radioactive waste treatment. A summary of the benefits and drawbacks of offered by these technologies is presented in Table 2.2.

Pre-ROD treatability studies provide critical performance and cost data (+50 percent to −30 percent accuracy) needed to evaluate and select an alternative for remedial action. However, the reliability and usefulness of the treatability study results are largely dependent on the chemical/physical characteristics of the wastes used in the conduct of the studies. The physical characteristics of the NFSS residues after removal from the WCS are not known at this time as the removal method has not been selected. Therefore, it is recommended that techniques for removing the residues from the WCS be evaluated prior to performing any treatability studies.

The actual K-65 residue sample volume that is available from FEMP for NFSS treatability studies is now limited to less than 5 kg. Obtaining this sample would require a formal request from the USACE to DOE and agreements would need to be negotiated regarding which agency maintains liability for transportation and management of the sample and disposal of the treatability study waste. These agreements are likely to be time consuming and complex to negotiate. Based on this information, conducting treatability studies to evaluate treatment technology performance for the residues does not appear to be possible at this time due to the unavailability of a representative sample (either from Silo 1 at FEMP or from the WCS).

At FEMP, the K-65 material has been declared 11e(2) material not regulated by the NRC and exempt from RCRA regulations. As a result, FEMP's focus has shifted from treatment of residues for reduction of toxicity and mobility to meet RCRA requirements and more towards treatment to cost effectively meet U.S. DOT requirements. Thus, it was assumed that the NFSS residues can be managed the same way since they are also 11(e)2 material not regulated by NRC.

If at the time the NFSS residues are removed, if removed at all, there are no commercial facilities available to handle the residues, then the only other management option would be disposal at a DOE facility. If the residues are disposed of at a DOE facility treatment would be required to meet the disposal facility WAC (which includes meeting the RCRA requirements). Therefore, at least one treatment technology that has the potential to reduce the toxicity and mobility of the residues to a level that meets RCRA requirements should be evaluated in the FS.

Due to the fact that treatability data is available from the FEMP treatment evaluations and the unavailability of a sample of residue material, it is concluded that treatability studies are not required prior to proceeding with the NFSS FS. Treatability data is available from FEMP at various scales (for bother surrogate and actual material) for vitrification, polymer based

encapsulation and chemical stabilization technologies (with numerous treatment reagents and chemical formulations). The FEMP treatability studies evaluated the ability to achieve immobilization of RCRA metals and reduction of direct radiation fields (to evaluate U.S. DOT compliant transportation and worker safety issues). This information can be used for the FS alternatives analysis and cost estimate. Resource recovery can be evaluated as metal recovery processes are proven and routinely applied. Existing characterization data from Fernald K-65 material can allow theoretical calculation of the economic viability of recovery.

It is recommended that although the FS can proceed without treatability studies, USACE should place themselves and the Program in a position where they can be responsive to any potential new technologies that may emerge during the CERCLA actions at NFSS and be able to integrate these new technologies or new treatability needs into any decisions regarding remediation. Therefore, USACE efforts should begin immediately to establish a cooperative agreement with the DOE for retrieval and use of adequate sample volumes to support the treatment technologies identified in this report and allow the opportunity to obtain additional sample volume should the need arise. Completion of the identified treatability studies would serve, at relatively low cost, to confirm the conclusions being formulated for the FS. In addition, such studies would likely provide valuable design data for full-scale remedial action.

# 6.0 REFERENCES

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# Appendix A Overview of Transmutation

The process of changing one element into another is transmutation. In 1939, Bohr and Wheeler theorized that energetic photons or gamma rays could produce fission, thus transmuting one element into two others. This was later shown to be true. For fissile materials, this means that gamma rays could induce fission, producing energy in a similar manner to a current nuclear power reactor. Fission frequently produces much shorter-lived products than the parent compounds.

Most radioisotopes are not fissionable in the sense of using thermal or higher energy neutrons to induce the reaction. Some,  $(U^{238}, Th^{232}, Pa^{231})$  however, can be made fissionable with higher energy (~1 MEV) neutrons. Non-fissionable isotopes can undergo another reaction which is labeled  $(\gamma,n)$ . This indicates that a gamma ray has induced the emission of a neutron. When this happens, the element remains the same, but the isotope changes. For example,  $Ra^{226}$  would become  $Ra^{225}$ . This could be considered a type of transmutation. Providing the energy of the gamma ray is correct – this type of reaction can be induced in most nuclei. Brown (unknown date) indicates that the process will work for elements from tritium (produces deuterium, which is stable) to  $Lead^{210}$  (produces  $Lead^{209}$  with a three hour half-life). According to Nuclear Solutions, Inc., the process is also applicable to  $Ra^{226}$ . Nuclear Solutions, Inc. holds the patent on an electron Accelerator-Driven System and is currently conducting research to apply their technology on spent nuclear fuel to transmute the long-lived isotopes into shorter-lived isotopes.

The residues stored at the NFSS WCS contain relatively high levels of  $Ra^{226}$ ,  $Pb^{210}$ ,  $Po^{210}$ , and  $Th^{230}$ . None of these are particularly fissionable; however, they are susceptible to the  $(\gamma,n)$  reaction. The determining factors are the binding energy of neutrons in the nucleus and the photonuclear cross section. A number of researchers in addition to Brown have investigated the process. In his paper on photoremediation, Brown cites Matsumoto as having calculated the effects on spent fuel rods containing the fission products  $Cs^{137}$  and  $Sr^{90}$ . His conclusion was that no pre-separation of the fusion products was needed to adequately spent fuel. Another researcher (Kase et.al., 1992) ran photofission feasibility experiments for  $U^{235}$ ,  $U^{238}$ ,  $NP^{237}$ , and  $Pu^{239}$ .

To date no pilot plant studies have been conducted to induce photoremediation (transmutation). The plant would need to include a number of pieces of equipment that are not normally thought of as remediation equipment. A linear accelerator accelerates electrons that are directed onto a high Z target such as tungsten to generate gamma rays with an energy of about 10 MeV. These gamma rays are directed onto the target material such as Ra<sup>226</sup> or U<sup>238</sup>. An accelerator of this type has been in operation in Japan since 1996. In addition to the reactor, a system for introducing the material to be irradiated into the beam and for handling and storage afterwards is needed. A method of dissipating or using the waste heat from the accelerator would also be needed. Additional equipment to monitor the process would be necessary so that the feed rates are optimized for transmutation.

The photonuclear data necessary to evaluate the full applicability of this technology to the NFSS residues was not readily available. Nuclear Solutions, Inc. has recently obtained the necessary data and SAIC is awaiting their input. A separate addendum to this report that outlines in greater detail a potential treatability study will be prepared once additional information is obtained from Nuclear Solutions, Inc.

Brown, Paul, unknown date. Photoremediation - An Emerging Treatment Technology. Nuclear Solutions, Inc.