INNOVATIVE TECHNOLOGY
Summary Report

DOE/EM-0471

Mercury Contamination – Amalgamate (contract with NFS and ADA)

Demonstration of DeHg^{SM} Process

Mixed Waste Focus Area

Prepared for

U.S. Department of Energy

Office of Environmental Management
Office of Science and Technology

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OST Reference #1675

Mixed Waste Focus Area

Demonstrated at
Pacific Northwest National Laboratory
Richland, Washington

Purpose of this document

Innovative Technology Summary Reports are designed to provide potential users with the information they need to quickly determine if a technology would apply to a particular environmental management problem. They are also designed for readers who may recommend that a technology be considered by prospective users.

Each report describes a technology, system, or process that has been developed and tested with funding from DOE's Office of Science and Technology (OST). A report presents the full range of problems that a technology, system, or process will address and its advantages to the DOE cleanup in terms of system performance, cost, and cleanup effectiveness. Most reports include comparisons to baseline technologies as well as other competing technologies. Information about commercial availability and technology readiness for implementation is also included. Innovative Technology Summary Reports are intended to provide summary information. References for more detailed information are provided in an appendix.

Efforts have been made to provide key data describing the performance, cost, and regulatory acceptance of the technology. If this information was not available at the time of publication, the omission is noted.

All published Innovative Technology Summary Reports are available on the OST Web site at http://OST.em.doe.gov under "Publications."

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SUMMARY

Technology Description

Through efforts led by the Mixed Waste Focus Area (MWFA) and its Mercury Working Group (HgWG), the inventory of bulk elemental mercury contaminated with radionuclides stored at various U.S. Department of Energy (DOE) sites is thought to be approximately 16 m^3 (Conley et al. 1998). At least 19 different DOE sites have this type of mixed low-level waste in their storage facilities.

The U.S. Environmental Protection Agency (EPA) specifies amalgamation as the treatment method for radioactively contaminated elemental mercury. Although the chemistry of amalgamation is well known, the practical engineering of a sizable amalgamation process has not been tested (Tyson 1993). To eliminate the existing DOE inventory in a reasonable timeframe, scalable equipment is needed that can

- produce waste forms that meet the EPA definition of amalgamation,
- produce waste forms that pass the EPA Toxicity Characteristic Leaching Procedure (TCLP) limit of 0.20 mg/L,
- limit mercury vapor concentrations during processing to below the Occupational Safety and Health Administration's (OSHA) 8-h worker exposure limit (50 $\mu g/m^3$) for mercury, and
- perform the above economically.

Additional major test objectives were to determine the mercury vapor pressure above the product and to assess the resistance of the treated wastes to degradation in a broad pH range.

Nuclear Fuel Services, Inc. (NFS) was selected to perform an amalgamation demonstration using their proprietary DeHg^{SM} (de´-merk) process. DeHg is a process capable of converting mercury-containing mixed waste of various matrices and chemical species to nonhazardous final waste forms. NFS performed the demonstration in its Applied Technology Laboratories in Erwin, Tennessee, under the 40 CFR 261.4 treatability exemption, which permits up to 1,000 kg of hazardous waste or 10,000 kg of mixed media to be tested. The demonstration apparatus and setup are shown in Figures 1 and 2.

Figure 1. Photograph of equipment based in a ventilation hood for the DeHg process.

Figure 2. Photograph showing close view inside hood of DeHg process.

DeHg was developed to treat mercury mixed waste containing not only elemental mercury, but also ionic and complexed forms of mercury. Based on the results of this demonstration on DOE wastes, adjustment of the DeHg process to address the chemistry of the mercury-contaminated matrix may be feasible. DeHg could also possibly be used to convert wastes bearing other codes commingled with D009, such as D005 (Ba), D006 (Cd), D008 (Pb), and D011 (Ag), to nonhazardous waste forms.

DeHg can be either a single- or two-step process, depending on the degree of difficulty in obtaining final-waste-form stability. In the first step, wastes are treated using classical amalgamation to stabilize elemental mercury contained in the waste. In the optional second step, supplemental amalgamating and/or stabilizing reagents are used to increase the stability of the final waste form. DeHg may be operated as either a “slurry” process for amalgamation/stabilization of shreddable debris wastes or strictly as an amalgamation process for bulk elemental mercury wastes. The process may also be modified for extraction of mercury from nonshredable wastes, soils, and sediments. More information about the process has been reported elsewhere (Davis 1998).

Demonstration Summary

In previous years, several treatability studies and other development efforts have been performed throughout the DOE complex related to amalgamation of mercury wastes. Such studies have used various materials to stabilize mercury. However, until this project was initiated, no studies beyond bench scale had been conducted. Consequently, the primary technical issue associated with the amalgamation of mixed waste mercury was related to scale-up of the process to a cost-effective operations level. For this reason, the HgWG issued a Request For Proposal (RFP), MER01, to industry in November 1996 entitled “Demonstration of the Amalgamation Process for Treatment of Radioactively Contaminated Elemental Mercury Wastes” (Simpson 1996).

The MER01 RFP sought to demonstrate the technical feasibility of and acquire engineering data for using amalgamation to treat radioactively contaminated bulk elemental mercury at DOE sites. One vendor selected was NFS, located in Erwin, Tennessee. NFS received bulk elemental-mercury mixed waste streams from three different sites for the amalgamation demonstration.

The DeHg process as applied to elemental mercury consists of two steps. The amalgamation unit operation addresses the elemental forms of mercury within the waste. The second chemical processing unit operation is used, if necessary, to stabilize mercury compounds or complexed forms of mercury. The solid waste form produced by this treatment train is leach tested and then either disposed (if it passes TCLP) or passed back to the amalgamation step (if it does not pass TCLP). Any liquid remaining after stabilization is either recycled or treated before disposal.

Following the two-step treatment with the DeHg process, samples of the amalgamated material were forwarded to Core Laboratories for Utah-certified analyses. According to certified laboratory assays, the amalgamated final waste forms not only achieved all applicable land disposal restrictions (LDR) and Utah criteria but also achieved <0.02 mg/L mercury in TCLP leachate, bettering the Universal Treatment Standard (UTS) limit of 0.025 mg/L for mercury. Details of analytical results are provided elsewhere (NSF 1998).

Key Results

All final waste forms appeared to be acceptable for disposal at the Envirocare site. The HgWG-advised conservative leaching standard for mercury (UTS of 0.025 mg/L, based on promulgation of new LDR limits) was also met in many instances.

The key results of the demonstration are as follows:

- met LDR and, in some cases, UTS for the radioactively contaminated mercury wastes processed,
- achieved mercury waste loadings of 20 to 25 wt %,
- used ambient-temperature processing to minimize mercuric oxide formation,
- is readily scalable to easily match the treatment needs at individual DOE sites,
- satisfies the EPA’s definition of an amalgam, as given in 40 CFR 268.42, Table 1, meeting disposal requirements outlined in the Resource Conservation and Recovery Act (RCRA).

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SECTION 2

TECHNOLOGY DESCRIPTION

Overall Process Definition

The EPA specifies amalgamation (AMLGM) as the best demonstrated available technology for the treatment of radioactively contaminated elemental mercury. EPA has defined AMLGM in Table 1 of 40 CFR 268.42 as

> Amalgamation of liquid, elemental mercury contaminated with radioactive materials utilizing inorganic agents such as copper, zinc, nickel, gold, and sulfur that result in a nonliquid, semi-solid amalgam and thereby reducing potential emissions of elemental mercury vapors to the air.

The DOE has in its legacy inventory approximately 16 m^3 of this type of waste in storage at 19 different sites. Demonstrations were performed with the following objectives in mind, as found in the Technology Development Requirements Document (TDRD) for mercury amalgamation (MWFA 1996), which can be found on the MWFA homepage waste.inel.gov/mwfa. For additional information, the personnel listed in the contacts list should be contacted.

- produce waste forms that meet the EPA definition of amalgamation,
- produce waste forms that pass the TCLP limit for mercury in nonwastewaters of 0.20 mg/L,
- limit mercury vapor concentrations during processing to below the OSHA 8-h worker exposure limit of 50 $\mu g/m^3$, and
- perform the above economically.

Additional major test objectives were to determine the mercury vapor pressure above the final waste form (amalgam) and to assess the resistance of treated wastes to a broad pH range. Personnel at the Oak Ridge National Laboratory (ORNL) will measure these two sets of parameters, and the results will be published separately.

A simplified block flow diagram of the NFS two-stage operation is provided in Figure 3. The overall approach is to first sort, shred, and slurry waste (if necessary) to create a homogeneous mixture and then to conduct chemical processing operations. The amalgamation unit operation addresses the elemental forms of mercury within the waste. If necessary, a second operation is added to stabilize mercury compounds or complexed forms of mercury.

The treated material emerges from the process as a presscake, which is then assayed for TCLP mercury and other metals. If the material passes, it becomes a candidate for further waste form profiling for ultimate disposal. If the material fails the criterion for leachable mercury, it may be reprocessed. The filtrate from the process is assayed for mercury and uranium. This water (filtrate) may be either recycled to the batch reactor for reuse or discharged.

Waste feed to the treatment system (Figure 3) would include feed from various sources, as shown in Figure 4. The most obvious source would be inventories already existing at user sites across the DOE complex. Other sources include generation from treatment processes for mercury or mercury-containing wastes. For example, elemental mercury may be derived as a product of retorting high-mercury (>260 ppm) wastes or recovered from the off-gas of a thermal treatment unit. This mercury must be amalgamated before it can be disposed. In each case, the waste would be characterized at the site (where it is in inventory or the point of generation) before packaging for shipment to the treatment facility. After processing at the treatment facility, the amalgamated, stabilized waste form would be tested to ensure that it meets the Waste Acceptance Criteria (WAC) of the intended disposal site. The final waste form would then be packaged and shipped for final disposition as low-level waste.

Description of Waste

NFS received streams from the following three sites for the amalgamation demonstration:

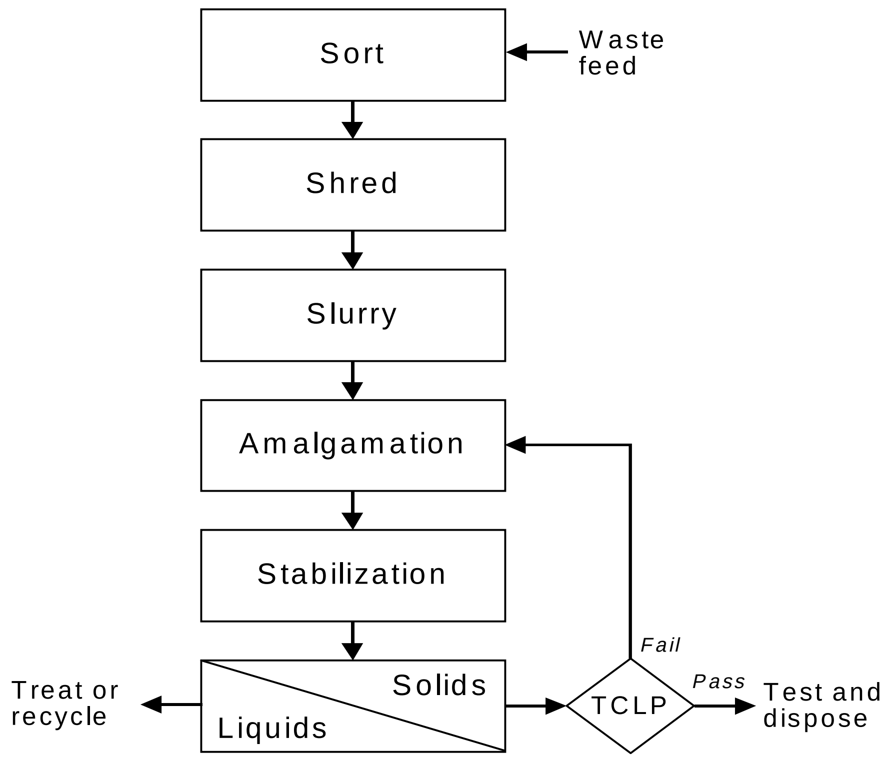

Figure 3. Block flow diagram of the DeHg mercury treatment process.

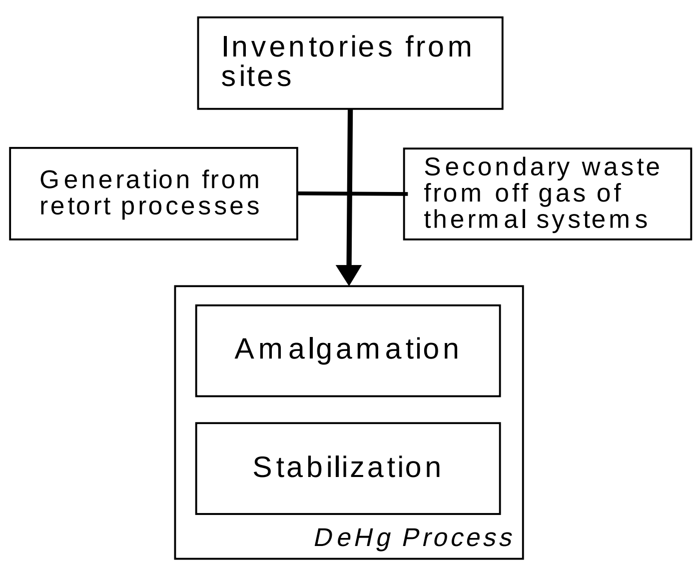

Figure 4. Sources of waste feed for the DeHg mercury amalgamation treatment process.

- 51 kg from the East Tennessee Technology Park (ETTP), formerly the K-25 Site;
- 23 kg from INEEL, operated by Lockheed Martin Idaho Technologies Company (LMITCO); and
- 1 kg from Diversified Scientific Services, Inc. (DSSI).

In total, NFS received 75 kg of test material from the three sites. The DSSI material was characterized as mercury that had been recovered from a thermal desorption treatability study on mercury-contaminated low-level radioactive soil from the INEEL.

System Operation

The DeHg process is operated at ambient temperature and pressure in a fully enclosed, ventilated hood. The process consists of two treatment steps preceded by a set of pretreatment operations for wastes other than the bulk elemental mercury. The first treatment step is an amalgamation technique to stabilize any elemental mercury the waste may contain, using one or more of the amalgamating agents as defined in 40 CFR 268.42. The second treatment step is a chemical stabilization process using a proprietary reagent to break mercury complexes and allow for removal of the mercury from the waste slurry as a stable precipitant.

Treatment campaigns may entail significant efforts dedicated to sorting material into similar matrices for assay and processing. Material suspected to be nonhazardous is removed at this stage and analyzed by TCLP protocol. Materials known to be contaminated with mercury or verified as hazardous by TCLP assays are sent to the treatment stage.

Containers emptied of the sample are soaked in a concentrated solution of a commercially available abrasive cleaner containing sodium hypochlorite. The containers are then rinsed with water, and a cleaning brush is used to remove any oil adhering to the sides. The containers were disposed as secondary waste.

Additional details, from sample preparation to wastewater management, are provided in Appendix B.

SECTION 3

PERFORMANCE

According to the strategy shown in Figure 5, three waste streams (see Table 1) were treated through the application of the DeHg process. In practice, the DSSI/INEEL stream was processed along with the LMITCO/INEEL stream. The performance results for the three streams are detailed in the two discussions below.

Processing INEEL Mercury

Elemental Mercury Phase

Mercury leachate concentrations for the initial 15 tests from first-step amalgamation averaged 1.2 mg/L and ranged from 0.05 to 7.5 mg/L. These values were above the regulatory limit of 0.2 mg/L mercury in TCLP leachate. Varying degrees of “oily sheen” were noted in the test aliquots. NFS developers theorized that oil entrained in the mercury phase was forming organomercury complexes, thereby resisting stabilization. Based on this theory, the amalgams were submitted to a second step of the DeHg process, which applies supplemental amalgamating/inorganic reagents to effect complete stabilization of mercury mixed wastes.

Table 1. Summary of radioactively contaminated elemental mercury waste streams provided to NFS for treatment in demonstration of the DeHg process.

Site	Quantity (kg)	Waste stream Characteristics
LMITCO/INEEL	23	Contains oil at 17% by volume[a] D009
LMES/ETTP	51	U 151[b]
DSSI/INEEL	1	Thermal Desorption Recovered Mercury D009

[a]NFS. 1998. *Demonstration of the DeHg[SM] Amalgamation Process for Radioactively Contaminated Elemental Mercury Wastes from Three Sites,* Draft Report submitted to Mixed Waste Focus Area, Mercury Working Group, by Nuclear Fuel Services, Inc., April 1998.

b. RCRA listed waste code for mercury.

Table 2 contains the results of both test series with and without the second step. The average process-control TCLP leachate mercury concentration of the two-step amalgams was 0.05 mg/L; TCLP leachate mercury concentrations ranged from 0.02 to 0.12 mg/L. These values were below the regulatory limit of 0.20 mg/L and in four cases were below the UTS limit of 0.025 mg/L for constituent concentrations in waste extract from the TCLP test.

The product of the combined DeHg process was comprised of polyethylene reaction chambers containing moist, semisolid amalgam without freestanding liquid. The 15 amalgams weighed a total of 114 kg.

In addition to the elemental mercury separated from the oil, a 13-mL specimen of clean elemental mercury was tested for treatability using the DeHg process. This sample of mercury had been recovered by DSSI (Kingston, Tennessee) from a sample of INEEL waste. The TCLP leachate mercury concentration from this material was 0.03 mg/L. No pretreatment of this mercury was required.

Oil Phase

The NFS analysis of TCLP leachate mercury concentration for this treated residue was 0.03 mg/L.

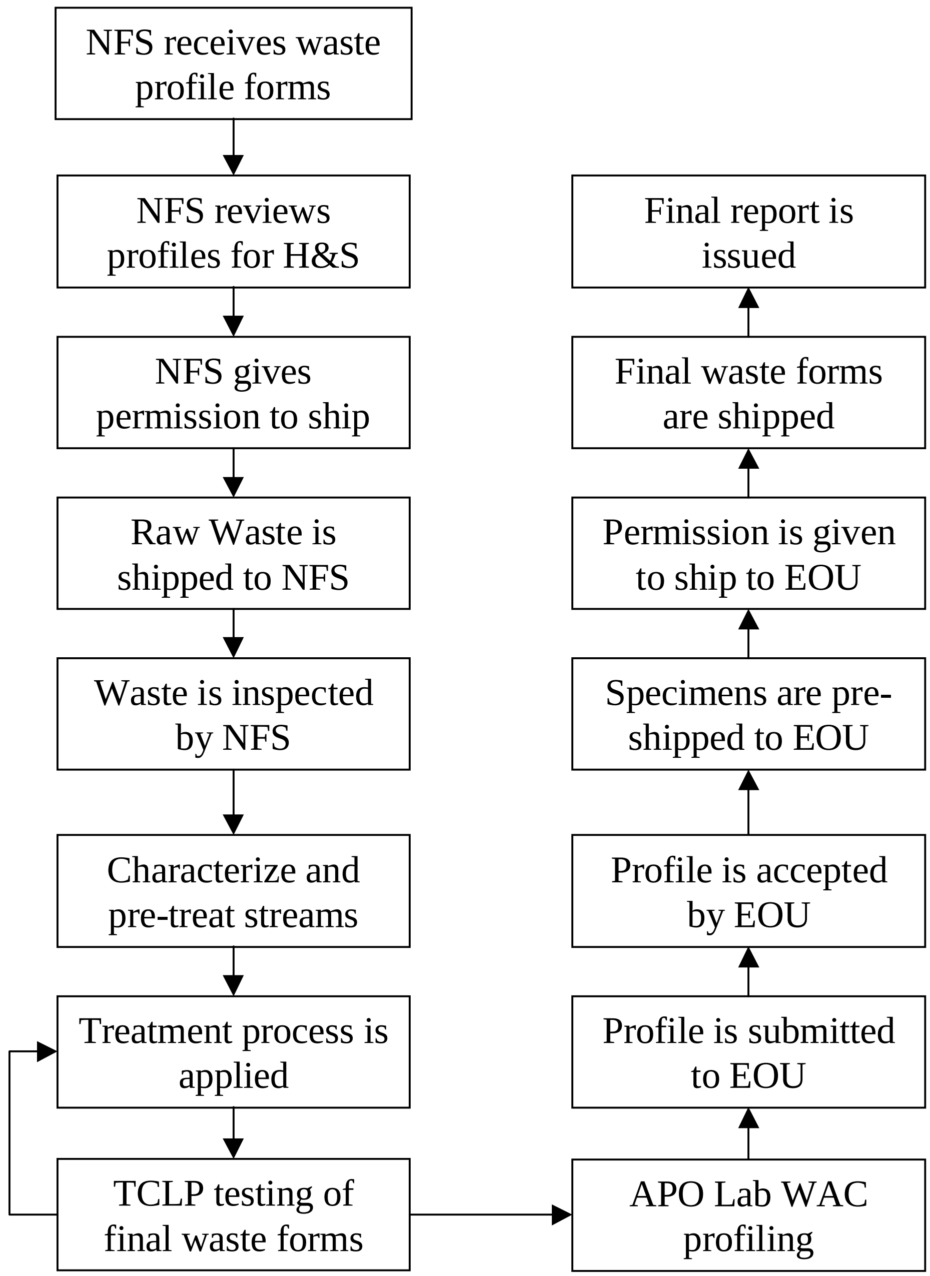

Figure 5. General flow logic for DeHg process demonstration.

Certified Assays of Amalgams, Residues, and Secondary Waste Streams

Table 3 shows the results of the TCLP mercury analyses performed at NFS, as well as the results furnished by Core Laboratories for the aforementioned samples. The results of the other State of Utah certification analyses are available elsewhere (NFS 1998).

Table 2. NFS process control data from the INEEL mercury amalgamation study.

Demonstration batch No.	One-step amalgamation TCLP Hg (mg/L)	Two-step Amalgamation TCLP Hg (mg/L)
1	0.31	0.03
2	0.05	NT[a]
3	0.44	0.07
4	0.11	NT
5	0.43	0.04
6	0.50	0.05
7	0.38	0.03
8	0.25	0.03
9	0.22	0.06
10	0.52	0.022
11	0.27	0.04
12	2.3	0.019
13	0.86	0.12
14	7.5	0.024
15	4.3	0.021

[a]Not tested.

Table 3. Tabulation of NFS and Sample Management Office-approved, Utah-certified laboratory assays

Sample description	TCLP [Hg], NFS (mg/L)	TCLP [Hg], Core Laboratories (mg/L)
Oil phase	150	50
Amalgam composite, sample 1	0.041	<0.02
Amalgam composite, sample 2	0.046	<0.02
Amalgam oil/Hg, sample 1	0.03	<0.02
Amalgam oil/Hg, sample 2	N/A	<0.02
"Clean" trash	0.077	<0.02
"Dirty" trash	0.054	<0.02

Table 4 shows the summary of all Utah-certified assays for the seven streams. As indicated by the Core Laboratories radioanalytical data (NFS 1998), the final INEEL waste forms contained radioactivity levels for uranium iso00topes and fission products that were generally in the <10-pCi/g range, which is well within disposal site acceptance limits.

Summary of Demonstration on INEEL Stream

The application of DeHg to the INEEL material produced amalgamated final waste forms that achieved criteria for disposal of material at Envirocare. NFS process control assays indicated an average TCLP leachate mercury concentration of 0.05 mg/L for all amalgam specimens based on small sample sizes. In four cases, the TCLP leachate mercury concentrations were below the UTS limit of 0.025 mg/L. Offsite assays performed by Core Laboratories indicated <0.020 mg/L for TCLP leachate mercury concentrations, which are below the EPA regulatory limit of 0.20 mg/L and the UTS regulatory limit of 0.025 mg/L.

Table 4. Sample Management Office (SMO)-approved, Utah-certified laboratory assays of INEEL mercury waste forms.

Parameter	Amalgam	Amalgam Duplicate	Stabilized oil	Oil Duplicate	Clean trash	Dirty Trash
Hg mg/L, TCLP	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
Pb mg/L, TCLP	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Ba mg/L, TCLP	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Cr mg/L, TCLP	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
As mg/L, TCLP	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Se mg/L, TCLP	<0.1	<0.1	0.1	1.2	<0.1	<0.1
Cd mg/L, TCLP	<0.01	<0.01	<0.01	<0.01	0.11	<0.01
Ag mg/L, TCLP	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Zn mg/L, TCLP	<0.1	<0.1	176	<0.5	10.9	67.9
Cu mg/L, TCLP	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Reactivity sulfide, mg/kg	50	20	<10	<10	20	<10
Reactivity cyanide, mg/kg	<10	<10	<10	<10	<10	<10
Paint filter test	Pass	Pass	Pass	Pass	2[a]	2[a]
Corrosivity (pH)	9.8	10.2	10.1	10.1	8.8	9.8
VOAs, μg/L TCLP	BQL[b]	BQL	BQL	BQL	BQL	BQL
Semi-VOAs, μg/L TCLP	BQL	BQL	BQL	BQL	BQL	BQL

[a]No free liquid noted by inspection.
[b]Below quantification limits.

The oily portion of the waste was also successfully treated using DeHg stabilization technology. The TCLP leachate mercury concentration for this residue was 0.03 mg/L; Core Laboratories assays indicated < 0.020 mg/L for TCLP leachate mercury concentrations, which is below the UTS limit for mercury of 0.025 mg/L.

Processing ETTP Mercury

Elemental Mercury Phase

Aliquots of the elemental mercury were treated using the first step of the DeHg process. The TCLP leachate concentrations for the first six batches averaged 0.29 mg/L mercury and ranged from 0.12 to 0.70 mg/L. Some of these values were above the regulatory limit of 0.20 mg/L mercury. NFS theorized that mercury species other than elemental mercury were not being stabilized in the amalgamation process. Based on this theory, the amalgams that were above the regulatory limit were submitted to the second step of the DeHg process. The balance of ETTP elemental mercury was also submitted to the two-step treatability demonstration, resulting in a total of 20 batch runs.

The product of the combined DeHg process consisted of polyethylene bottles of moist amalgam with no freestanding liquid. The 20 bottles weighed a total of 238 kg. The average TCLP leachate mercury concentrations of the amalgams were 0.05 mg/L and ranged from 0.01 to 0.17 mg/L. These values were comfortably below the regulatory limit of 0.20 mg/L and, in two cases, were below the UTS limit of 0.025 mg/L.

Certified Assays of Amalgams and Secondary Waste Streams

Table 5 shows the results of the TCLP mercury analyses performed at NFS, as well as the results furnished by Core Laboratories for the aforementioned samples. Table 6 shows the summary of all Utah-certified assays for the three streams.

Table 5. Tabulation of NFS and Utah-certified laboratory assays.

Sample description	TCLP [Hg], mg/L NFS	TCLP [Hg], mg/L Core Labs
Amalgam composite, sample 1	0.12	<0.02
Amalgam composite, sample 2	0.05	<0.02
Trash	NT[a]	<0.02

[a]Not tested.

Table 6. Utah-certified laboratory assays of ETTP mercury waste forms.

Parameter	Amalgam	Amalgam duplicate	Trash
Hg mg/L, TCLP	<0.02	<0.02	<0.02
Pb mg/L, TCLP	<0.2	<0.2	<0.2
Ba mg/L, TCLP	<0.01	<0.01	0.07
Cr mg/L, TCLP	<0.05	<0.05	<0.05
As mg/L, TCLP	<0.2	<0.2	<0.2
Se mg/L, TCLP	<0.2	<0.2	<0.2
Cd mg/L, TCLP	<0.02	<0.02	<0.02
Ag mg/L, TCLP	<0.03	<0.03	<0.03
Zn mg/L, TCLP	61.1	23.7	<0.1
Cu mg/L, TCLP	<0.05	<0.05	<0.05
Reactivity sulfide, mg/kg	<10	<10	<10
Reactivity cyanide, mg/kg	<10	<10	<10
Paint filter test	pass	pass	Pass
Corrosivity (pH)	9.7	9.8	10.4
VOAs, μg/L TCLP	BQL[a]	BQL	BQL
Semi-VOAs, μg/L TCLP	BQL	BQL	BQL

[a]Below quantification limit.

As indicated by the Core Laboratories radioanalytical data for the ETTP material, the final waste forms provided have radioactivity levels for uranium isotopes and fission products that are in the <10-pCi/g range, which is well within disposal site acceptance limits.

Summary of Demonstration on ETTP Stream

NFS amalgamated ETTP elemental mercury to produce final amalgams having TCLP leachate mercury concentrations that averaged 0.05 mg/L, according to process control assays. These results were safely below the EPA regulatory limit of 0.20 mg/L.

In two cases, the TCLP leachate mercury concentrations were below the UTS limit of 0.025 mg/L. All samples analyzed at an independent offsite laboratory had TCLP leachate mercury concentrations <0.02 mg/L, which were below the UTS limit of 0.025 mg/L.

SECTION 4

TECHNOLOGY APPLICABILITY ALTERNATIVES

Competing Technologies

Baseline Technologies

RCRA regulations call for amalgamation of elemental mercury waste that cannot be recycled; this category includes radioactive elemental mercury. Hence, amalgamation is the baseline process for treatment of elemental mercury waste. Amalgamation is the process of alloying mercury with another metal, such as zinc, copper, nickel, gold, or sulfur, to form a combination that is solid at room temperature. In waste treatment, the objective of amalgamation is the stabilization of mercury for subsequent separation or disposal. The primary type of waste that requires amalgamation is free elemental mercury, although a few waste streams with treatability matrix parameters of debris and soil have been slated for amalgamation treatment by DOE sites.

Mercury amalgamation has been used throughout modern history to extract precious metals (e.g., gold, silver) from metal ore. RCRA regulations impose requirements for the final waste form, which must also be met through adherence to the UTS or a defined technology-based treatment standard. Only in the relatively small-scale dental applications are there commercial uses of amalgamation technology to produce waste forms with requirements similar to those in waste management.

Various treatability studies and other development efforts performed throughout the DOE complex have explored the ability of several different materials to stabilize mercury, including tin, zinc, copper, sulfur, and sulfur polymer cement. However, until this demonstration, known investigations on mixed waste amalgamation were limited to bench scale. Hence, there is no commercial-scale technology available to serve as a baseline.

Other Competing Technologies

A *Commerce Business Daily* announcement (Request For Information) by the HgWG on mercury treatment technology capabilities produced 42 responses. Among the respondents, all of whom received the subsequent RFPs, only two proposals pertained to amalgamation. The RFP responses may be taken as a major indicator of existing capabilities across the nation and of vendors desiring to prove their technology. Both bids were found to be acceptable by the HgWG, based on its pass/fail criteria (MWFA 1996). From these two bids, demonstration contracts were put in place with the respective vendors: one to NFS for its process and another to ADA Technologies, Inc. (ADA, Englewood, Colorado). Hence, the ADA process is seen as the only one competing with that of NFS. As there is no appropriate baseline, these two competing technologies will be compared.

ADA proposed a process for stabilizing radioactively contaminated elemental mercury with sulfur. The process combines a proven mercury stabilization method with a scalable, economically viable mixing technology. In the ADA process, waste mercury is mixed with sulfur in a commercially available pug mill, producing a stable mercury sulfide product. The pug mill is well-suited to the process because of its ability to adequately mix the components and control the residence time to ensure complete reaction. In addition, radioactive contamination control requirements, necessary for dealing with mixed waste, can be readily implemented using the pug mill.

The findings from the ADA demonstration are reported in detail in a separate Innovative Technology Summary Report (ITSR). The product from the ADA demonstration passed TCLP treatment standards and met vapor pressure requirements (during processing) described in the TDRD. The process also satisfies the EPA's definition of an amalgam, as given in 40 CFR 268.42, Table 1, satisfying disposal requirements as outlined in RCRA regulations.

Technology Comparison

The comparison of the NFS process with the competing amalgamation process technology demonstrated by ADA is shown in Table 7. In general, applications of the NFS process will require a two-step version of the process: amalgamation plus further stabilization. An important limiting factor is the high cost. Although the NFS process is estimated to cost less than the ADA process, the cost is high.

The treatment of mercury wastes requiring amalgamation will be very expensive unless additional economies of scale can be capitalized upon. Larger-scale processing operations, such as a centralized facility, will tend to be favored economically over multiple smaller, site-based operations.

Technology Applicability

The NFS process is specifically suited to the treatment of mercury waste, including existing streams and secondary waste streams of elemental mercury produced from thermal systems used to treat mercury-contaminated wastes. As noted earlier, pretreatment steps may be required in the treatment train.

Table 7. Comparison of ADA and Nuclear Fuel Services, Inc. (NFS), AMALGAMATION TECHNOLOGIES.

Comparison factor	NFS	ADA
Pretreatment methods	None for elemental mercury	None for elemental mercury
Process mechanism	Standard laboratory glassware operation based in a ventilation hood	Pug mill, a dual-shaft mixer, with liner to decrease dead volume
Particle size of the amalgamating material	Processes are similar in this regard; particle sizes are on the order of 50–100 microns	
Control of free mercury in the final waste	Both forms eliminated free mercury in the final waste	
Optimal waste loading	20–25%[a]	50–60%
Final waste form	Passes TCLP and, largely, UTS	Passes TCLP
Amalgamation process	Uses proprietary additives and EPA-prescribed agents	Uses sulfur and a small amount of proprietary liquid
Effect of contaminants on the process	Tolerated oily phase in Hg	Water <10% tolerated; other contamination not addressed
Definition of amalgamation	Both processes meet the definition of amalgamation	
Amalgam waste loading	20–25%[a] achieved	57% achieved in demonstration
Formation of mercuric oxide	Both processes employ low (ambient) operating temperatures and were easily able to meet OSHA requirements, minimizing these concerns	
Formation of mercury vapor		
Secondary waste streams	Bottles, solutions, gloves, tissues, rags, and lab coats are 15% of final waste form	Sand, personal protection equipment, decontamination materials, and filters amount to about 5% of stabilized waste
Decomposition of final waste	Being evaluated at ORNL	Being evaluated at ORNL
Ambient environmental conditions	Both processes are operated at ambient conditions	
Process temperature		
Control of mercury vapor	Process operated in a ventilated hood	Forced convection of room air through a high-efficiency particulate air (HEPA) filter
Moisture	Moisture (water in small amounts) is tolerated by both processes	
Throughput	80 kg/8 h at full scale	>100 kg/8 h at full scale
Duty cycle	Both processes designed to operate an 8-h shift, 5 days/week	
Reliability	Both processes use commercially available equipment and proprietary reagents; demonstrated reliability and maintainability are similar	
Maintainability		
Transportability	Both processes could be deployed as mobile units; equipment is small enough to fit in a truck; minimal utility requirements	
Physical characteristics	Waste form characteristics are physically similar	
Waste Acceptance Criteria	Both processes produce waste forms that meet current Envirocare Waste Acceptance Criteria (WAC)	
Regulatory and safety requirements	No additional hazards, safety, or regulatory issues found for either process	
Public and tribal involvement	The processes are identical in this respect	
Quality assurance and testing	Both processes were demonstrated according to an MWFA-approved Quality Assurance Plan	
Disposition of equipment and waste	Both processes are similar in this regard	
Estimated cost (for 1,000–1,500 kg)	Slightly lower than ADA cost, but volume considerations are key	Slightly higher than NFS cost, but volume considerations are key
Summary assessment	Less costly; better leach performance (in ability to meet UTS)	Higher waste loadings, less secondary waste, and fewer proprietary reagents

[a]NFS stated an exception to this criterion that the loading of mercury in its bulk elemental mercury amalgamation process was in the 20 to 25 wt% range based on performance criteria needed for LDR.

SECTION 5

COST

Methodology

As part of the original Statement of Work (SOW) (Simpson 1996), the HgWG requested an evaluation of detailed life-cycle cost estimates for processing of radioactively contaminated mercury wastes. An engineered facility exists at the NFS facility. Hence, NFS addressed this requirement by providing an estimate of the price to process material based on the waste stream characteristics encountered in this demonstration (MWFA 1996). This price is based on the projected NFS facility rate estimated below.

Estimation Basis

The permitted maximum daily rate (24 h) for DeHg amalgamation of bulk elemental mercury is 768 kg. This rate is achieved by the simultaneous operation of multiple batch units. The estimated rate at 62% capacity is 160 kg/8 h for one-step processing. If the UTS criterion is desired for the final waste form, then a two-step process is required, for which the effective rate is estimated to be 80 kg/8-h shift. This rate is in excess of the minimum rate of 45.5 kg/8 h specified in the original SOW (Simpson 1996). The amalgamation system rate may be turned up or down in virtually any mass increments to address treatment loading. For example, if necessary, a rate as low as 1 kg/h or less may be used (MWFA 1996).

Based on the elemental mercury specimens used in this demonstration, a generic flow sheet and associated material balance were developed. This information was presented earlier in Figure 6. The flow sheet shows material flows with a basis of 100 kg of total input, with all processing performed at ambient conditions. If the feed input contains oil, as shown in the diagram, a phase separation is invoked as pretreatment. The two phases, elemental mercury and oil, are then routed as separate streams for amalgamation of the elemental mercury and stabilization of the oil phase. In this flow diagram, the reagent dosage for oil stabilization is estimated and is not intended to be an optimized value. For the case of bulk mercury not containing commingled oil, the phase separation may not be required and direct amalgamation of mercury occurs.

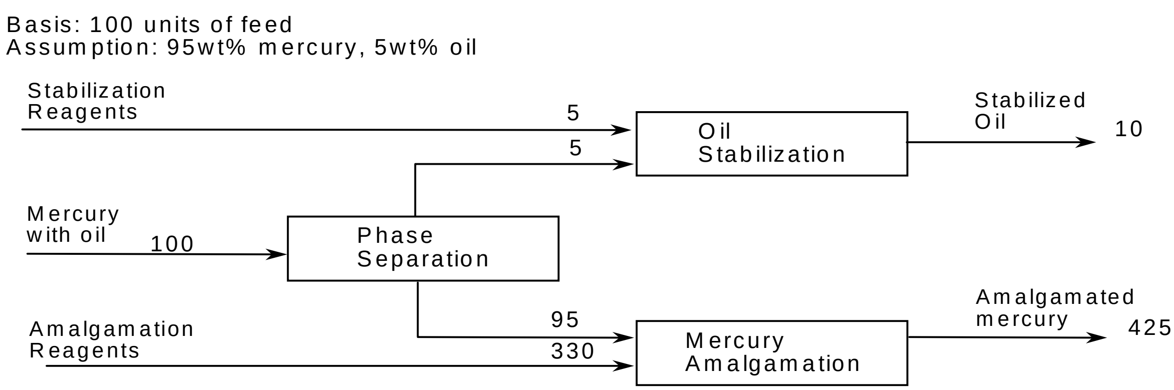

Figure 6. Simplified flow sheet for DeHg amalgamation process applied to INEEL stream.

The NFS site is assumed to be the location for processing of the bulk elemental mercury for the purpose of this estimate.

The estimated pricing provided by NFS does not include transportation and burial, but includes waste profiling assays by a Utah-certified laboratory. The estimate does not address batch volumes larger than 1,500 kg and is expected to change if the DeHg system is deployed at a Treatment, Storage, and Disposal Facility (TSDF).

Although NFS possesses a permitted facility, NFS is not a TSDF. NFS has indicated that they have considered options to deploy their DeHg amalgamation technology at other TSDF sites that are licensed and permitted for mixed waste treatment. NFS continues to look for off-site deployment opportunities, presenting the potential for cost efficiencies in terms of reduced site and labor overhead.

Cost Conclusions

In prior efforts, the HgWG has found that the major obstacle to deploying mercury-treatment technologies is the small quantities of most waste types at a given site. A single technology capable of treating most of the streams would be prohibitively expensive to deploy. Cost estimates for treatment of wastes from a single site at an offsite vendor are also seen to be extremely high. These findings are true for amalgamation based on information and costs estimates supplied by ADA and NFS. For this reason, national contracts combining the wastes of a given type from the DOE complex under a single treatment contract offer a rational, cost-effective approach to treatment technology deployment. The HgWG has planned a national contract for treatment of elemental mercury waste. Further information about national contract planning by the HgWG is provided in Appendix B. To avoid jeopardizing this national initiative, the exact vendor cost figures will not be published in this report. However, both vendors indicated that if either were to be contracted to treat over 1,500 kg of elemental mercury, the cost would be approximately $300/kg. This estimate assumes that the waste is elemental mercury that can be treated in one large production run without interim system decontamination requirements. Disposal costs of the treated waste are not included in the estimate. The treatment of mercury wastes requiring amalgamation will be very expensive, unless additional economies of scale can be capitalized on. Larger-scale processing operations, such as a centralized facility, will tend to be favored economically over multiple smaller, site-based operations.

SECTION 6

REGULATORY AND POLICY ISSUES

Regulatory Considerations

The regulatory/permitting issues related to the use of amalgamation technology are governed by the following safety and health regulations:

- Occupational Safety and Health Administration (OSHA), 29 CFR 1926
  - 1926.28 Personal Protective Equipment
  - 1926.102 Eye and Face Protection
  - 1926.103 Respiratory Protection
- OSHA 29 CFR 1910
  - 1910.132 General Requirements (Personnel Protective Equipment)
  - 1910.133 Eye and Face Protection
  - 1910.134 Respiratory Protection.

Disposal requirements/criteria include the following Department of Transportation and DOE requirements:

- 49 CFR, Subchapter C, Hazardous Materials Regulation
  - —171 General Information, Regulations, and Definitions
  - —172 Hazardous Materials Table, Special Provisions, Hazardous Materials Communications, Emergency Response Information, and Training Requirements
  - —173 Shippers—General Requirements for Shipments and Packagings
  - —174 Carriage by Rail
  - —177 Carriage by Public Highway
  - —178 Specifications for Packaging
- 10 CFR 71 Packaging and Transportation of Radioactive Material.

If the waste is determined to be hazardous solid waste, the following EPA requirement should be considered:

- 40 CFR, Subchapter 1 Solid Waste.

CERCLA Criteria

The Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) has established nine criteria against which alternative treatment approaches are to be judged during the Remedial Investigation/Feasibility Study (RI/FS) portion of the remediation action. A short explanation of each of the criteria (EPA 1988) and the assessment of the NFS process against it follows.

Overall protection of human health and the environment
This criterion is an evaluation of the overall protectiveness of an alternative. The criterion focuses on whether a specific alternative achieves adequate protection and describes how site risks posed through each pathway being addressed by the feasibility study (FS) are eliminated, reduced, or controlled.

In a CERCLA environment, the resulting waste forms from the NFS process will provide improved protection of human health and the environment by reducing the mobility of the elemental mercury. The amalgams are placed inside a plastic container to further enhance protection.

Compliance with ARARs
This evaluation criterion is used to determine whether each alternative will meet all of the federal and state Applicable or Relevant and Appropriate Requirements (ARARs) that have been identified in previous stages of the RI/FS process.

The Land Disposal Restrictions are the most likely ARARs to be applied to a CERCLA site dealing with elemental mercury. These regulations under RCRA specify amalgamation for elemental mercury. The NFS process meets that definition.

Long-term effectiveness and permanence
Alternatives under this criterion are to be evaluated in terms of risk remaining at the site after response objectives have been met. The primary focus of this evaluation involves the extent and effectiveness of the controls that may be required to manage the risk posed by treatment residuals and/or untreated wastes.

The long-term effectiveness of any remediation process has to be judged not only by the efficacy of the actual treatment process, but also by how well the process can be applied to the extent of the contamination. Assuming that the elemental mercury can be efficiently brought to NFS equipment, the process should be able to provide environmental protectiveness. Tests to be performed at ORNL will provide a more definitive answer.

Reduction of toxicity, mobility, or volume through treatment
The statutory preference for selecting remedial actions that employ treatment technologies that permanently and significantly reduce toxicity, mobility, or volume of the hazardous substances is to be evaluated under this criterion.

Amalgamation should significantly reduce the mobility of mercury in a waste management scenario. In a CERCLA action, further study would be required to assess how the action of bacteria affects the waste form. Secondary containment is prudent in any case.

Short-term effectiveness
This criterion addresses the effects of the alternative during the construction and implementation phase until remedial response objectives are met.

As designed and operated, the NFS process should be protective of the community and the workers while not imposing meaningful environmental consequences during its operation.

Implementability
The implementability criterion focuses on the technical and administrative feasibility of implementing an alternative and the availability of various services and materials required during implementation.

The process should prove to be viable from the standpoints of both the technical (ability to construct, reliability, and monitoring) and administrative (coordination with other agencies) feasibility, as well as the availability of services and materials.

Cost
The costing procedures found in the *Remedial Action Costing Procedures Manual* are to be the bases for comparing alternatives with regard to costs.

The cost figures to be provided in the future were not based on the rigorous details provided in the referenced document above.

State acceptance
This assessment evaluates the technical and administrative issues and concerns the state (or support agency in the case of state-lead sites) may have regarding each of the alternatives.

See the section entitled “Safety, Risks, Benefits, and Community Reaction."

Community acceptance

Under this criterion, an assessment is made on the issues and concerns the public may have regarding each of the alternatives.

See the following section.

Safety, Risks, Benefits, and Community Reaction

Following the completion of their laboratory development phase, NFS assembled a project team to complete the system design and develop the requisite licenses and permits for operating the system. The NFS design team was multidisciplined, composed of chemists, engineers, health physics personnel, nuclear safety engineers, and regulatory specialists to address key issues concerning chemical safety, radiological control, permitting, and licensing issues. NFS found no special issues with respect to the hazards associated with their process. Various aspects of risk have been evaluated by the HgWG, considering eight criteria for the level of risk as associated with mercury amalgamation, as follows:

1. correctness (technical correctness),
2. cost (effectiveness to use),
3. permittability (ease of permitting),
4. safety,
5. sponsorship (commitment by sponsors),
6. completeness (ready for use),
7. acceptability (to stakeholders), and
8. timeliness (to meet schedules).

The risk values, established for the MWFA-developed technology processes, have been derived from top-level requirements defined in the MWFA Systems Requirements Document. Evaluations of the technology and assignment of risk values were made by a team comprised of HgWG members, in consideration of the risk category definitions and performance observations from the demonstration experience. The assessments made are summarized below.

Correctness

This risk category is rated as very low. The targeted volume of waste to be treated is small as compared with most other waste types. The fact that amalgamation is required by law reflects the fact that this type of technology is appropriate for this waste. The performance demonstrated by NFS adequately addresses any concerns over the capability of their process to successfully treat mercury mixed waste.

Cost

This risk category is rated as moderate. The targeted volume to be treated is low, but the waste possesses diverse characteristics. Oxidation, complexation, and speciation of mercury across various matrices add an element of uncertainty as to the difficulty of successfully stabilizing the bulk of inventory (and future generation) without process modifications. In addition, cost estimates provided by NFS are characterized by some uncertainty.

Permittability

This risk category is rated as very low. The treatment process is simple and based on well-proven Best Demonstrated Available Technology for nonradioactive mercury waste. The volumes of waste involved are small enough to pose little likelihood of regulatory problems.

Safety

This risk category is rated as low. While mercury is a hazardous material of some concern and radioactive contamination has the potential to raise additional concern, mercury vapors and leaching appear to be well controlled by the process and radioactive contamination is low. The stability of the final waste form is dominant in immobilizing both mercury and radionuclides, thereby minimizing concerns over worker safety, public safety, and environmental protection.

Sponsorship

This risk category is rated as moderately low. Interest by the sites has been good, and programmatic support for technology development has demonstrated good commitment. There is a small risk that some potential users may find a local or on-site solution for treatment of their mercury wastes.

Completeness

This risk category is rated as moderately low due to the simple, proven nature of amalgamation. The potential complexity of the chemistry of mercury and the diversity of waste matrices adds to the risk for the system.

Acceptability

This risk category is rated as very low. Amalgamation is a process easily identifiable to the public because of its long-time use by the dental profession. The waste form stability, simplicity, and small-scale nature that characterize the technology are expected to make for easy public acceptance.

Timeliness

This risk category is rated low. Based on preliminary information received to date from 10 DOE sites, the timeframe for treatment is late FY 1999 and FY 2000. If a national contract is put in place, all sites will have a reliable route to use for disposal of their mercury wastes requiring amalgamation, via a single vendor under contract.

Public Participation

The siting of a mixed waste treatment facility of any type near communities will involve public input. Stakeholders are generally concerned about the type, toxicity, and amount of emissions to be discharged to the atmosphere and the disposal site for the final waste form.

The MWFA Tribal and Public Involvement Resource Team and HgWG initiated activities to involve and gather stakeholder issues, needs, and concerns about mercury treatment technologies. These activities included reviews, articles, and presentations. During November and December 1997, the chair of the HgWG addressed both the Oak Ridge Local Oversight Committee and the Site-Specific Advisory Board (SSAB). The purpose of the meetings on November 17–18, 1997, was to identify issues, needs, and concerns of various Oak Ridge stakeholders regarding technologies that may be applicable to Oak Ridge. The areas emphasized included continuous emission monitors, characterization, input to Technology Performance Reports, and the HgWG. These meetings were interactive, where participants explored the issues and problem solved collectively. No formal presentations were made, but information was provided and progress on various MWFA projects was discussed. Participants included members of the local oversight committee, the Site Technology Coordination Group, and the general public.

The SSAB Environmental Technology Group meeting on December 10, 1997, involved providing stakeholder input into various technology development projects at Oak Ridge. Those they have expressed interest in addressing are:

- Transportable Vitrification System,
- Toxic Substances Control Act (TSCA) Test Bed for Continuous Emissions Monitors,
- Mercury Working Group/Mercury Treatment Demonstrations, and
- Removal of Mercury from Liquid Wastes.

A short presentation on the status of each activity was given, and the proposed future scopes were discussed.

The MWFA assembled a Technical Requirements Working Group (TRWG), which is a stakeholder group capable of representing varied tribal and public perspectives. The TRWG assisted MWFA technical staff in transforming or integrating site-specific issues, needs, and concerns into the TDRDs and in providing tribal and public perspectives to technical staff for identifying and resolving technical issues. The TRWG reviewed and provided recommendations to the MWFA on changes to the Mercury Amalgamation TDRD.

Lastly, the MWFA Resource Team facilitated tribal and public involvement by issuing an article in the quarterly newsletter (July 1997) highlighting mercury treatment and disposal.

The plan for a national contract for mercury waste treatment is consistent with minimizing concerns over siting for what would otherwise be multiple treatment facilities. While there are still transportation issues, these are expected to be routine and to present no special concerns. No tribal issues are anticipated.

SECTION 7

LESSONS LEARNED

Implementation Considerations

The lessons learned from this process showed no surprises in waste handling or processing conditions required to adequately treat the waste.

Technology Limitations and Needs for Future Development

The high cost of treatment indicates a need for cost reduction measures such as those that could be gained through the implementation of a national procurement contract.

Some unknown factors still exist that pertain to the effects of contaminants in commercial-scale amalgamation of mercury-contaminated mixed wastes. Also, the effect of speciation has been explored only minimally. Available information is limited, and further investigation is needed.

Technology Selection Considerations

The process, as demonstrated, works well on elemental mercury. When the process is located at a facility with a RCRA Part B permit, the primary consideration will be the extent of cocontamination of both RCRA substances and radionuclides.

APPENDIX A

References

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APPENDIX B

TMS Data Elements

Sample Preparation

Waste samples are separated into physical phases before characterization and subsequent treatment.

Characterization

Grab sampling of waste streams and substreams was performed in the demonstration. Total-mercury and TCLP analyses were performed by NFS before treatment to serve as a baseline for treatment performance and to provide data needed to determine reagent dosages for the chemical stabilization step. Additional posttreatment analyses were performed by Core Laboratories (Casper, Wyoming).

Amalgamation of Elemental Mercury

The amalgamation procedure was performed on the waste. Specific compositions and reagent quantities were determined based on waste inspection and characterization results. The procedure is described in more detail elsewhere (NFS 1998).

Stabilization of Solid- and Aqueous-Phase Substreams

Solid and aqueous substreams that failed to meet TCLP criteria for mercury were treated.

Stabilization of Secondary Solid Waste

Any secondary waste that became D009 was treated and shipped with the treated material to the waste disposal site (Envirocare).

Wastewater Management

Wastewater, including rinse water, generated during processing was collected and analyzed for mercury and radioactive constituents. Wastewater exceeding permitted release limits was treated to remove target contaminants to meet discharge limits. Wastewater that met discharge limits was processed for discharge under National Pollutant Discharge Elimination System permit guidance.

Waste Form Testing

Analytical testing was performed to determine if TCLP limits were met. Determinations of percentage residual free mercury and mercury vapor pressure were also important performance measurements.

Material and Energy Requirements

Material requirements include amalgamation and stabilization reagents. Analytical and testing equipment is required for the characterization and TCLP procedures.

Energy requirements are nominal, consisting simply of what is required for pumping, mixing, analysis, and testing.

Skill and Training Requirements

The operational procedures require no special skills beyond those of a chemical operator or technician. Oversight of the operation requires a project chemist or chemical engineer. Compositional analysis and leach-testing procedures can be performed by a technician. However, the results should be interpreted and certified by a chemist or engineer.

The operator must be trained in the operating procedures and should be familiar with the acceptance criteria of the target disposal site, but no additional special training is required.

APPENDIX C

ACRONYMS

ADA	ADA Technologies, Inc. (Englewood, Colorado)
APO	Analytical Projects Office
ARARs	Applicable or Relevant and Appropriate Requirements
BOIP	Balance of Inventory Procurement (i.e., Broad Spectrum Procurement)
CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act
CFR	Code of Federal Regulations
DeHg	Proprietary process by NFS for processing Mercury mixed waste (pronounced de´-merk)
DOE	Department of Energy
DSSI	Diversified Scientific Services, Inc.
EOU	Envirocare of Utah
EPA	Environmental Protection Agency
ETTP	East Tennessee Technology Park
FS	Feasibility Study
HgWG	Mercury Working Group, MWFA
H&S	Health and Safety
INEEL	Idaho National Engineering and Environmental Laboratory
ITSR	Innovative Technology Summary Report
LDR	Land Disposal Restrictions
LMITCO	Lockheed Martin Idaho Technologies Company
MER01	Solicitation to industry (November 1996) entitled “Demonstration of the Amalgamation Process for Treatment of Radioactively Contaminated Elemental Mercury Wastes”
MWFA	Mixed Waste Focus Area
NFS	Nuclear Fuel Services, Incorporated (Erwin, Tennessee)
ORNL	Oak Ridge National Laboratory
OSHA	Occupational Safety and Health Administration
ppm	parts per million
RCRA	Resource Conservation and Recovery Act
RFP	Request For Proposal
RI/FS	Remedial Investigation/Feasibility Study
SOW	Statement of Work
SSAB	Site-Specific Advisory Board
TCLP	Toxicity Characteristic Leaching Procedure
TDRD	Technology Development Requirements Document
TRWG	Technical Requirements Working Group
TSDF	Treatment, Storage, and Disposal Facility
UTS	Universal Treatment Standard
WAC	Waste Acceptance Criteria