Technical Report

Advances in Encapsulation Technologies for the Management of Mercury-Contaminated Hazardous Wastes

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26 W. Martin Luther King Drive
Cincinnati, OH 45268

Paul M. Randall
Task Order Project Officer

Prepared by

Sandip Chattopadhyay
Wendy E. Condit

Battelle
505 King Avenue
Columbus, OH 43201-2693

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Abbreviations and Acronyms

ANL	Argonne National Laboratory
ANS	American Nuclear Society
ANSI	American National Standard Institute
ARD	acid rock drainage
AS_2O_3	arsenic trioxide
ASTM	American Society for Testing and Materials
BDAT	best demonstrated available technology
BNL	Brookhaven National Laboratory
BP	bench-scale/pilot-scale
CBPC	chemically bonded phosphate ceramics
CFR	Code of Federal Register
CSF	ceramic silicon foam
DOE	United States Department of Energy
DOT	United States Department of Transportation
F	full-scale
Fe_2O_3	haematite
H_2S	hydrogen sulfide
HDPE	high-density polyethylene
Hg_2SO_4	mercurous sulfate
$HgCl_2$	mercuric chloride
HgS	mercuric sulfide
INEL	Idaho National Engineering Laboratory
KH_2PO_4	monopotassium phosphate
LDPE	low-density polyethylene
LDR	Land Disposal Restrictions
MA	macroencapsulation
MgO	magnesium oxide

MI	microencapsulation
NA	not applicable
Na_2S	sodium sulfide
$Na_2S.9H_2O$	sodium sulfide nonahydrate
Na_2SO	sodium sulfite
NR	not reported
NRC	Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
ppm	parts per million
psi	pounds per square inch
RCRA	Resource Conservation and Recovery Act
SAIC	Science Application International Corporation
SEM	scanning-electron microscope
SITE	Superfund Innovative Technology Evaluation
SPC	sulfur polymer cement
STC	Silicate Technology Co.
TCE	trichloroethylene
TCLP	Toxicity Characteristic Leaching Procedure
U.S. EPA	United States Environmental Protection Agency
VOC	volatile organic compound
vol%	volume percent
wt%	weight percent
WPI	Waste Policy Institute

Abstract

Although industrial and commercial uses of mercury have been curtailed in recent times, there is a demonstrated need for the development of reliable hazardous waste management techniques because of ongoing hazardous waste generation and historic operations that have led to significant contamination. The focus of this article is on the current state of encapsulation technologies and materials being used to immobilize elemental mercury, mercury-containing debris, and other mercury-contaminated wastes, soils, or sludges. The range of encapsulation materials used in bench-scale, pilot-scale, and full-scale applications for mercury-containing wastes are summarized in this report. Several studies have been completed regarding the application of sulfur polymer stabilization/solidification, chemically bonded phosphate ceramic encapsulation, and polyethylene encapsulation. Other technologies or materials reported in the literature or under development for encapsulation include asphalt, polyester resins, synthetic elastomers, polysiloxane, sol-gels (e.g., polycerams), and Dolocrete™. The objective of these encapsulation methods is primarily to physically immobilize hazardous wastes to prevent contact with leaching agents such as water. These methods may also include a stabilization step to chemically fix mercury into a highly insoluble form. Economic information relating to the use of these materials is provided, along with available vendor information. Future technology development and research needs are also discussed.

1.0 Introduction

The development of effective treatment options for mercury-contaminated wastes is a significant technical and practical challenge due to several factors, including the limited economic benefit derived from mercury recovery/recycling, the high toxicity, volatility, and environmental mobility of mercury, and the varied nature and composition of industrial waste products. Principal sources of mercury-contaminated industrial wastes include chloralkali manufacturing, weapons production, copper and zinc smelting, gold mining, paint applications, and other processes (United States Environmental Protection Agency [U.S. EPA], 1997). Although industrial and commercial uses of mercury have been curtailed in recent times, there is a demonstrated need for the development of reliable hazardous waste management techniques because of ongoing hazardous waste generation and historic operations that have led to significant contamination. This document focuses on the current state of encapsulation technologies and materials being used to immobilize elemental mercury, mercury-containing debris, and mercury-contaminated wastes, soils, and sludges.

As an inorganic element, mercury cannot be destroyed, but it can be converted into less soluble or leachable forms to inhibit migration into the environment after disposal. The management and ultimate disposal of mercury-contaminated hazardous waste is controlled by U.S. EPA regulations known as the Land Disposal Restrictions (LDRs) (40 Code of Federal Register [CFR] Part 268). Under the current LDR program, the U.S. EPA has established thermal recovery (e.g., roasting/retorting) as the best demonstrated available technology (BDAT) for treatment of wastes containing greater than 260 mg/kg of mercury. For treatment of wastes with less than 260 mg/kg of mercury, other extraction technologies (e.g., acid leaching) or immobilization technologies (e.g., stabilization/ solidification) may be considered (U.S. EPA, 1999). Because mercury contained in radioactive or mixed waste is not suitable for thermal recovery and recycling, the U.S. EPA also recognizes that stabilization/solidification may be an appropriate treatment option for heavily contaminated mercury mixed wastes or debris (Waste Policy Institute [WPI], 1999).

Stabilization/solidification relies upon mobility reduction resulting from a combination of chemical reaction (e.g., precipitation) and physical entrapment (e.g., porosity reduction). Encapsulation technologies are based primarily on solidification, and act to prevent hazardous waste from coming into contact with potential leaching agents, such as water. Hazardous waste materials can be encapsulated in two ways: microencapsulation or macroencapsulation. Microencapsulation involves mixing the waste together with the encasing material before solidification occurs. Macroencapsulation involves pouring the encasing material over and

around a larger mass of waste, thereby enclosing it in a solidified block. Sometimes these processes are combined.

The U.S. EPA is considering changes to the LDR program to require a macroencapsulation step prior to the land disposal of stabilized mercury wastes. Mercury wastes may be stabilized using sulfide or other chemical fixation processes, but the stabilization process is pH dependent and may not permanently immobilize mercury for disposal. The optimal pH range is 4 to 8 for chemical fixation of mercury compounds to the highly insoluble solid form, mercuric sulfide (HgS). At high pH, the more soluble solids mercurous sulfate (Hg_2SO_4), mercuric sulfate ($HgSO_4$), and mercury sulfide hydrogen sulfide complex ($HgS[H_2S]_2$) are formed depending on oxidizing or reducing conditions; while at low pH, hydrogen sulfide gas escapes from the waste (Wagh et al., 2000; Clever et al., 1985). Combining stabilization with macroencapsulation to prevent pH-related degradation of the treated waste may improve its long-term stability and therefore minimize any potential threats to human health and the environment.

The range of encapsulation materials used in bench-scale, pilot-scale, and full-scale applications are summarized in the following sections. Economic information for several different encapsulation materials is provided, along with available vendor information. Future technology development and research needs are also discussed.

2.0 Encapsulation Materials

Materials used for encapsulation of mercury must be both chemically compatible with the hazardous waste and inert to common environmental conditions that may be encountered in a disposal facility, such as rain infiltration, groundwater flow, and freeze/thaw cycles. Sulfur polymer stabilization/solidification (SPSS), chemically bonded phosphate ceramic (CBPC) encapsulation, and polyethylene encapsulation are just three of the techniques that are currently being tested and used to improve the long-term stability of hazardous wastes. Studies that focus on the management of elemental mercury, mercury-contaminated debris, and other mercury-contaminated wastes will be discussed. Each encapsulation material will be reviewed in terms of the key features of the encapsulation process, current applications and technology status, and available performance data. The advantages and disadvantages associated with each material will also be discussed.

Performance data for encapsulated wastes typically include both physical data (e.g., strength, density, and permeability) and/or chemical data (e.g., leachability). For macroencapsulated waste, the most important evaluation criteria are the compressive strength, the waste form density, the presence of void spaces, and the barrier thickness. The primary concern during macroencapsulation is that an inert surface coating or jacket is created which substantially reduces the potential for exposure of the waste to leaching media (Mattus, 1998). For microencapsulated waste, the toxicity characteristic leaching procedure (TCLP) plays an important role in determining whether or not the material can be accepted by a landfill. Macroencapsulated materials are typically not tested with the TCLP. According to the Resource Conservation and Recovery Act (RCRA) LDR rules, mercury hazardous waste is defined as any waste that has a TCLP value greater than 0.2 mg/L. Mercury-contaminated wastes that exceed this value must be treated to meet the Universal Treatment Standard (UTS) of 0.025 mg/L or less prior to disposal in a landfill. The TCLP test methodology, Method 1311, is described in detail in the U.S. EPA guidance document *SW-846 Test Methods for Evaluating Solid Waste, Physical/Chemical Methods*. TCLP tests on microencapsulated material may require size reduction to meet the particle size specifications in Method 1311. Instead of crushing, cutting, or grinding the microencapsulated material, the particle size requirements are typically met through subsampling the waste and binder formulation and casting small pellets in the appropriate size. This methodology helps to meet TCLP test requirements, while maintaining the barrier surface and integrity of the waste form. The U.S. Nuclear Regulatory Commission (NRC) has also developed its own waste form testing protocols for mixed wastes. In general, NRC waste form testing examines the influence of environmental factors on the final waste form stability including the effect of thermal cycling and immersion on compressive strength and the impact of

biodegradation and irradiation. However, a detailed discussion of these waste evaluation criteria are beyond the scope of this report.

2.1 Sulfur Polymer Stabilization/Solidification

Conventional stabilization/solidification methods typically include the fixation of metals using Portland cement and fly ash, which produces an impermeable, solid waste form and creates a high pH environment that limits the solubility and leachability of most metals. However, it is very difficult to stabilize mercury with cement-based processes because it does not form a low-solubility hydroxide solid (U.S. EPA, 1999). For this reason, a significant amount of research has gone into the development of alternative binding materials for the stabilization/solidification of mercury-contaminated wastes. As discussed below, the SPSS process can be used to both convert mercury compounds into the highly insoluble HgS form and to simultaneously encapsulate the waste.

The SPSS process relies upon the use of a thermoplastic material which contains 95 wt% elemental sulfur and 5 wt% of the organic modifiers, dicyclopentadiene and oligomers of cyclopentadiene. This material is referred to in the literature as sulfur polymer cement (SPC), although it is not a cementitious material. SPC melts at approximately 115°C (235°F) and sets rapidly upon cooling. It is relatively impermeable to water compared to conventional concrete and has a high mechanical strength at approximately double that of conventional concrete. SPC is also well suited to harsh environments with high levels of mineral acids, corrosive electrolytes, or salt solutions, according to research completed by van Dalen and Rijpkema (1989), McBee and Donahue (1985) and others as quoted in Darnell (1996).

Figure 2-1 provides a simplified block-diagram for the SPSS encapsulation process (United States Department of Energy [DOE], 1994). For macroencapsulation, molten SPC is poured over and around large debris such as metal scrap and is then allowed to set into a monolithic waste form. The recommended mixing temperature for SPC is between 127-138 ^{o}C (260-280 ^{o}F). Operating in this range will minimize gaseous emissions and provide optimum viscosity (Darnell, 1996).

For microencapsulation of liquid, elemental mercury, a two-stage process referred to as SPSS has been patented by Kalb et al. of Brookhaven National Laboratory (BNL) under U.S. Patent No. 6,399,849. First, the elemental mercury is mixed in a heated reaction vessel at 40 ^{o}C with powdered SPC. Other chemical stabilization agents such as sodium sulfide and triisobutyl phosphine sulfide can also be added during this initial step. The heated reaction vessel helps to accelerate the reaction between mercury, SPC, and the additives to form HgS and an inert gas atmosphere helps to prevent the formation of mercuric oxide. Next, additional SPC is added and the mixture is heated to 130 ^{o}C (266 ^{o}F) to form a homogenous molten liquid, which is then poured into a mold and allowed to set into a monolithic waste form. This two-step process minimizes both the oxidation of mercury to mercuric oxide and the amount of unreacted mercury. In addition, the researchers have confirmed the formation of two forms of mercuric sulfide as a result of the treatment process. Both meta-cinnabar and cinnabar phases were identified using x-ray powder diffraction scans (Fuhrmann et al., 2002). BNL has two patents related to sulfur polymer encapsulation (U.S. Patents No. 6,399,849 and 5,678,234). BNL recently has licensed the SPSS technology to Newmont Mining Corporation for the

encapsulation of liquid elemental mercury generated as a byproduct of gold mining operations. Newmont and BNL are currently working on scaling-up the technology for industrial use (BNL, 2002).

Several studies have been completed regarding the use of SPC for metal-contaminated wastes including Fuhrmann et al. (2002), Mattus (1998), and Darnell (1996). The results and observations from these studies are discussed below, along with a summary of the advantages and limitations associated with the SPC encapsulation method. Table 2-1 summarizes key performance data from these studies.

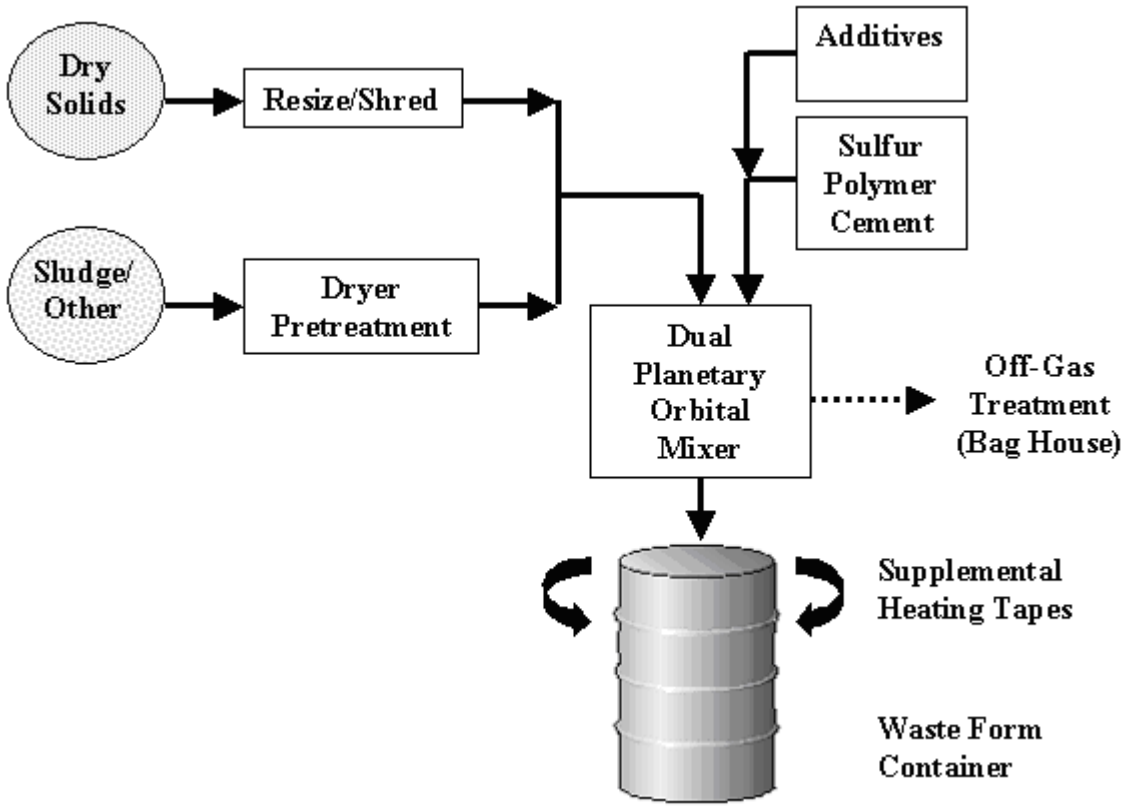

Figure 2-1. Sulfur Polymer Stabilization/Solidification

Note: Figure modified from DOE (1994). Additives can be used to decrease the leachability of mercury. Additives reported in the literature include sodium sulfide and tri-isobutyl phosphine sulfide (Fuhrmann et al., 2002).

Fuhrmann et al. (2002) presents the results from a bench-scale study for the treatment of radioactive elemental mercury with the patented SPSS process described above. Elemental mercury and radioactive elemental mercury were obtained from waste stocks at Brookhaven National Laboratory. The study explored three issues including the leachability of the treated waste, the formation of mercuric sulfide, and mercury vaporization during processing. Several treatability tests were conducted on the mercury wastes including microencapsulation with SPC alone, with 3 wt% triisobutyl phosphine sulfide, 3 wt% sodium sulfide nonahydrate, and a 1.5 wt% combination of these two additives. Microencapsulation of the elemental mercury with

SPC alone resulted in TCLPs ranging from 20 ug/L to > 400 ug/L. The final formulation that was chosen was the 3 wt% sodium sulfide treatment which resulted in an average leachate concentration for mercury of 25.8 ug/L and a range from 1.3 to 50 ug/L. Long-term leaching studies were also conducted according to the American Society for Testing and Materials (ASTM) Method C-1308. This test demonstrated a very low release rate with a diffusion coefficient for mercury in the final waste form on the order of 10^{-17} cm^2/s. The authors also explored the formation of mercuric sulfide through x-ray diffraction studies and determined that elemental mercury and SPC reacted to form primarily meta-cinnabar. However, elemental mercury and sodium sulfide nonahydrate formed primarily cinnabar, which explains the improved leaching behavior in those tests with 3 wt% sodium sulfide as an additive. The results of mercury volatilization studies also demonstrated that mercury volatilization was reduced through the treatment with sodium sulfide. Headspace measurements for elemental mercury ranged from 9.2 to 12.7 mg/m^3 in vapor, ranged from 0.41 to 4.5 mg/m^3 with just SPC, and 0.20 to 1.3 mg/m^3 with the addition of sodium sulfide. These results suggest that, for adequate retention of the mercury during processing, the use of additives such as sodium sulfide may be necessary (Fuhrmann et al., 2002).

Oak Ridge National Laboratory (ORNL) completed a treatability test to scale-up the SPC process for the macroencapsulation of mixed waste debris, contaminated with mercury and other metals (Mattus, 1998 and ORNL, 1997). The ORNL treatability study objectives included scaled-up equipment selection, determination of the size and shape of the final waste form, and process parameter monitoring and optimization. The treatability study was performed using two mixed waste streams generated at ORNL:

- 208 kg (457 lb) of cadmium sheets (Resources Conservation and Recovery Act [RCRA] waste code D006), and
- 204 kg (448 lb) of lead pipes contaminated with mercury (RCRA waste codes D008 and D009).

The cadmium sheets were classified as debris under the alternative debris standards found in 40 CFR 268.45. Macroencapsulation is an option for treatment of this waste code under the alternative debris treatment standards. Macroencapsulation with SPC also satisfied the LDR treatment standards for radioactive lead solids (D008) and for mercury (D009) through amalgamation, as sulfur is one of the elements that is able to form a non-liquid, semisolid when combined with mercury.

Table 2-1. Key Performance Data for Sulfur Polymer Stabilization/Solidification

Author/ Vendor	Type	Scale	Waste Type	Waste Form Size	Waste Loading (wt%)	Compressive Strength (psi)	Density (g/cm^3)	Before Hg TCLP (mg/L)	After Hg TCLP (mg/L)
Mattus (1998)	MA	BP	Mixed waste cadmium sheets	5 gal	15.8 to 28.6	NR	NR	NA	NA
Mattus (1998)	MA	BP	Mixed waste lead pipes/gloves contaminated with Hg	5 gal	31.3 t0 38.8	NR	NR	NA	NA
Fuhrmann et al. (2002)	MI	BP	Radioactive Hg^o	5 gal	33.3	NR	NR	2.64	0.020 to >0.40
Fuhrmann et al. (2002)	MI	BP	Radioactive Hg^o with 3 wt% triisobutyl phosphine sulfide additive to SPC	5 gal	33.3	NR	NR	2.64	> 0.40
Fuhrmann et al. (2002)	MI	BP	Radioactive Hg^o with 3 wt% $Na_2S.9H_2O$ additive to SPC	5 gal	33.3	NR	NR	2.64	0.0013 to 0.050
Darnell (1996)[(b)]	MI	BP/F	Metal oxides including Hg, Pb, Ag, As, Ba, and Cr at 5 wt% each	NR	40	4,000	NR	250[(a)]	<0.2
Kalb et al., (1996)	MI	BP	Mixed waste off-gas scrub solution	NR	25 to 45	3,850 to 8,160	1.86 to 1.94	0.14	<0.009

BP=Bench-Scale/Pilot-Scale.
F= Full-Scale.
MA= macroencapsulation
MI= microencapsulation,
NA= not applicable.
NR= not reported.
TCLP = Toxicity Characteristic Leaching Procedure.
(a) Untreated waste TCLP not reported, so estimated based on total Hg level in waste divided by 20.
(b) Sodium sulfide nonahydrate was added to reduce metal leachability.

The equipment used in the study included primarily a 5-gallon steel container, a rigid wire basket, a handle spacer to hold the debris in place, a vibrating table, external heater tapes, and a melting pot and pour pipe for the molten SPC. The size and shape of the final waste form was developed based upon criteria from the United States Department of Transportation (DOT) hazardous material shipping regulations (49 CFR 173.12), the hazardous waste disposal facility (Envirocare), and general safety and handling considerations. The DOT requirements included the use of an approved container with a total waste form weight limit of 205 kg. The Envirocare facility specified that the following process requirements would have to be met for waste acceptance:

- The barrier had to be in intimate contact with the waste,
- The barrier should be at least 2 inches thick around the waste material, and
- The waste had to be encapsulated using a continuous pour.

Because of the small size of the waste form mold, some preparation and size reduction of the cadmium sheet debris and lead pipe wastes was required. In a radiological fume hood, sheet metal scissors were used to reduce the size of the cadmium sheets, which ranged in length from 4 inches to more than 40 inches. For the lead pipe wastes, pieces of debris were bent or cut to the target size (e.g., < 4 inches).
The SPC process was first tested with non-contaminated materials, so the waste form could be cut transversely and observed to optimize process parameters. Following two practice trials, the radioactive mixed waste streams were encapsulated in a series of 20 batches. The major steps involved in the SPC macroencapsulation process included the following:

1) The debris was placed into a secured wire basket, which was centered in the drum to maintain a 2-inch surrounding layer of SPC,

2) Molten SPC was poured into the drum to provide both the outer layer of SPC and to fill the voids between the debris,

3) The pour was continued until SPC reached 2 inches from the top of the drum,

4) Once the bottom portion of the waste form had hardened, the spacer holding the wire basket was removed and a cap of molten SPC was added to fill the drum, and

5) Once the cap layer was set, the drum was sent for land disposal.

It was found that heating the debris to 140 to 150 ^{o}C (284 to 302 ^{o}F) for six hours prior to the pour helped to ensure that fast cooling of the SPC did not occur at the waste-binder interface and helped to reduce the formation of air pockets. Vibrating the container throughout the pouring sequence and for up to five minutes after the pour also improved setting of the waste form. Heating tapes were used to maintain a target temperature of 190 ^{o}C (374 ^{o}F) at the top portion of the container. This allowed air bubbles from the setting SPC to escape. The optimal additional heating time was determined to be 10 hours after the pour had ended. During the surrogate waste test, examination of the waste form cross section revealed good contact between the debris pieces and SPC and no identifiable interface between the two pour layers (i.e., the top portion of

the drum and the cap). No H_2S or SO_2 off-gasses were detected during the tests. For macroencapsulation of the mixed wastes, waste loadings for the cadmium sheets ranged from 15.8 to 28.6 wt% and for the lead pipes ranged from 31.3 to 38.8 wt%. Key performance data from this study is summarized in Table 2-1.

Darnell (1996) demonstrated the use of SPC for the microencapsulation of up to 5 wt% of metal oxides including mercury, lead, silver, arsenic, barium, and chromium. Darnell microencapsulated a variety of metal-contaminated wastes including dehydrated boric acid salts, incinerator hearth ash, mixed waste fly ash, and dehydrated sodium sulfate salts. These treated wastes were then subjected to the U.S. EPA TCLP, the Nuclear Regulatory Commission (NRC) waste form qualification testing, and the American Nuclear Society (ANS 16.1) leaching index test. Darnell also found that an additional chemical stabilization step was needed to treat mercury to meet TCLP limits. A 7 wt% sodium sulfide nonahydrate ($Na_2S.9H_2O$) was added to the SPC mixture to convert metal oxides to more leach-resistant metal sulfides. The U.S. EPA TCLP limits were achieved for all metals. Key performance data from this study is summarized in Table 2-1.

Darnell (1996) also discussed the issues considered during scale-up of the SPC encapsulation process for mixed waste incinerator fly ash. The full-scale system proposed consisted of a large disposal box (1 m on each side) that would be stacked in an above-grade, earth-mounded, concrete disposal vault. The box would be surrounded by a heated mold-form to prevent swelling due to the approximately 3,000 lb of waste and SPC to be placed inside. Both the box and the waste would be preheated to the melt temperature to prevent the SPC from freezing upon contact. Automated steam or oil-heated mixers were planned to provide temperature control and to allow the mixer to be shut down or restarted during a pour. Temperature controls for mixing and cooling would be computer controlled with the appropriate alarms for safety (e.g., gas detection alarms).

The following is a list of advantages and limitations associated with the use of SPC for the encapsulation of hazardous wastes:

Advantages

- SPSS results in the formation of a very insoluble sulfide compound with mercury (HgS).
- SPSS is well-suited to the treatment of elemental Hg.
- No chemical reaction is required for SPC to set and cure; therefore greater waste-to-binder ratios are allowed than with Portland cement.
- Relatively low temperature process (127-138 ^{o}C or 260-280 ^{o}F).
- Superior water tightness (e.g., low permeability and porosity) compared to Portland cement.
- High resistance to corrosive environments (e.g., acids and salts).
- SPC has a high mechanical strength.

- SPC is resistant to freeze-thaw cycling and has coefficients of expansion compatible with other construction materials.
- Simple to implement because mixing and pouring equipment is readily available.
- SPC is easier to use than other thermoplastics, like polyethylene, because of its low viscosity and low-melt temperature.
- It is possible to remelt and reformulate SPC.

Limitations

- Although SPC encapsulation occurs at relatively low temperatures, volatile losses of mercury may occur and engineering controls are needed. BNL's patented SPSS process was designed to minimize Hg volatilization and 99.7% of the Hg treated remains in the waste form, while only 0.3% of the Hg is volatilized and captured in an off-gas system.
- Aqueous wastes must be dewatered prior to processing.
- If cooled too quickly, SPC will develop an excess of voids or air pockets, which could allow water or gas to penetrate the waste form.
- Metal debris or pieces with large thermal mass may require debris preheating above the SPC melting point to prevent the formation of air pockets around the debris-binder interface.
- Not compatible with strong alkaline solutions (>10%), strong oxidizing agents, or aromatic or chlorinated solvents.
- Expanding clays cannot be used in SPC.
- SPC handling requires the use of engineering controls to mitigate possible ignition and explosion hazards.
- If excessive temperatures are created, SPC will emit hydrogen sulfide gas and sulfur vapor.

2.2 Chemically Bonded Phosphate Ceramic Encapsulation

Chemically bonded phosphate ceramics (CBPCs) are well suited for encapsulation because the solidification of this material occurs at low temperatures and within a wide pH range. The DOE Argonne National Laboratory (ANL) has six patents covering the use of this material for the encapsulation of hazardous wastes. The technology has been licensed to Wangtec, Inc., for the treatment of incinerator ashes from power plants in Taiwan (DOE, 1999a). Similar to the SPC technology, successful treatment with CBPC is due to both chemical stabilization and physical encapsulation. Although mercury will form low solubility phosphate solids, stabilization with a small amount of sodium sulfide (Na_2S) or potassium sulfide (K_2S) to form HgS greatly improves the performance of the final CBPC waste form. $Hg_3(PO_4)_2$ has a solubility product of 7.9×10^{-46}, compared to HgS with a solubility product of 2.0×10^{-49} (Wagh et al., 2000).

CBPCs are fabricated through an acid-base reaction between calcined magnesium oxide (MgO) and monopotassium phosphate (KH_2PO_4) in solution to from a hard, dense ceramic of magnesium potassium phosphate hydrate as shown in the reaction below:

$$MgO + KH_2PO_4 + 5\ H_2O \rightarrow MgKPO_4.6\ H_2O\ (MKP)$$

Iron oxide phosphates can also be used to form a low-temperature ceramic, but research into the use of this material is limited (Seidel et al., 1998). CBPC waste forms typically have a density of 1.8 g/cm^3 and high compressive strengths (>2,000 psi). They also have an open porosity that is up to 50% less than conventional fabricated cement. Waste loadings up to 78% have been demonstrated with this technology. Figure 2-2 provides a simplified block-diagram for the CBPC encapsulation process. First, enough water is added to the waste in the disposal drum to reach a target or stoichiometric water content. (One advantage of the CBPC process is that it can be carried out on dry solids, wet sludges, or liquid wastes.) Next, calcined magnesium oxide and monopotassium phosphate binders are ground to a powder and blended in a one-to-one ratio. Additional ingredients (e.g., fly ash or K_2S for mercury fixation) also are added to the binders. The water, binders, additional ingredients, and waste are mixed for about 30 minutes. Under most conditions, heat from the reaction causes the waste matrix to reach a maximum temperature of approximately 80 ^oC (176 ^oF). After mixing is stopped, the waste form typically sets in about 2 hours and cures in about two weeks. Mixing can be completed in a 55-gallon disposal drum with a planetary type mixer. The waste, water, binder, and additives can be charged to the drum using hoppers, feeding chutes, and piping as needed (DOE, 1999a).

Several detailed studies have been completed to demonstrate the use of CBPCs for both macroencapsulation and microencapsulation of hazardous wastes including Singh et al. (1998), DOE (1999a), Wagh et al. (2000), and Wagh and Jeong (2001). These papers describe the steps involved in fabricating the CBPC waste forms and also discuss the results of various performance tests including compressive strength measurements, U.S. EPA TCLP tests, and leaching index tests. Visual observations of the structural integrity of the waste forms were also made. The CBPC encapsulation process has been tested on a wide variety of hazardous wastes including low-level, mixed waste ash, transuranics, fission products, radon-emanating wastes, salt solutions, and heterogeneous mercury-containing debris (Wagh and Jeong, 2001). Table 2-2 summarizes key performance data from these studies.

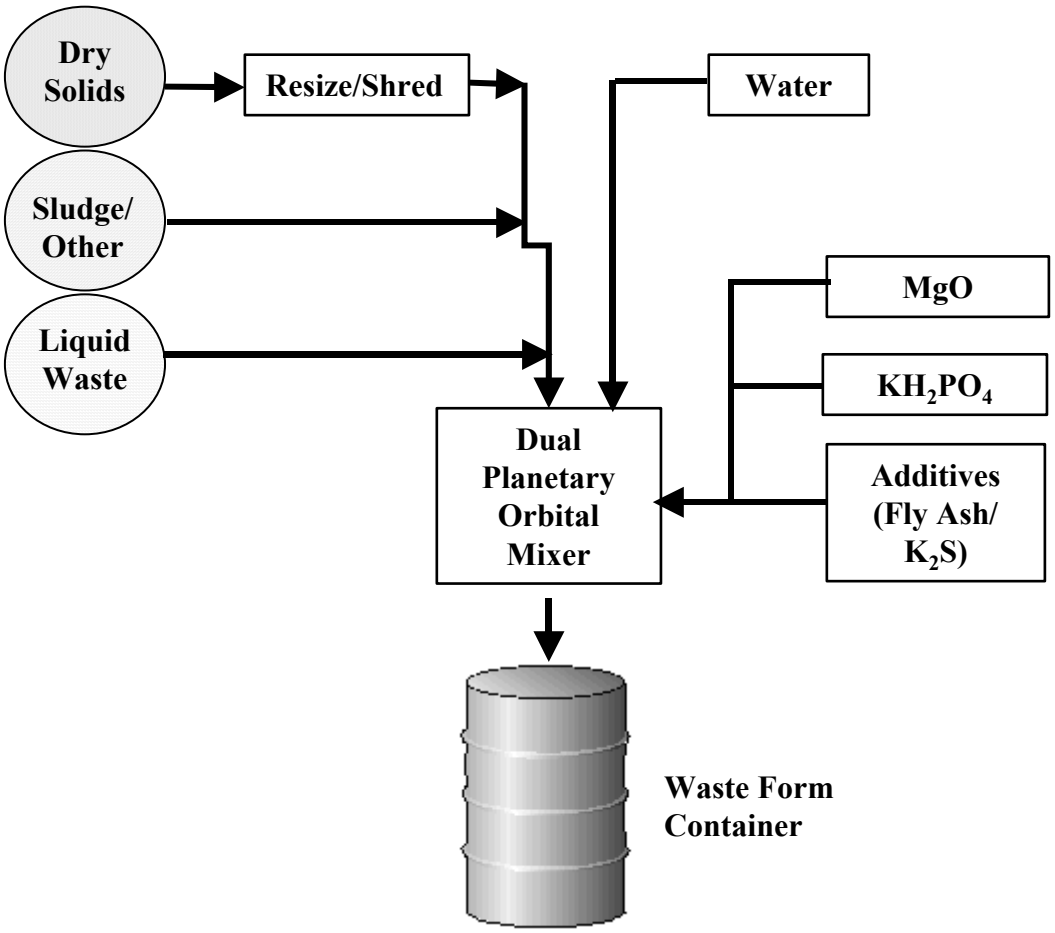

Figure 2-2. Chemically Bonded Phosphate Ceramic Process

Table 2-2. Key Performance Data for Chemically Bonded Phosphate Ceramic Encapsulation

Author/ Vendor	Type	Scale	Waste Type	Waste Form Size	Waste Loading (wt%)	Compressive Strength (psi)	Density (g/cm^3)	Before Hg TCLP, (mg/L)	After Hg TCLP (mg/L)
Singh et al. (1998)[b]	MA	BP	Cryofractured debris	1.2 to 3 gal	35	5,000 to 7,000	1.81	NA	NA
Singh et al. (1998)[b]	MA	BP	Lead bricks	NR	NR	5,000 to 7,000	1.8	NA	NA
Sing et al. (1998)[b]	MA	BP	Lead-lined gloves	5 gal	NR	5,000 to 7,000	1.8	NA	NA
Singh et al. (1998)[b]	MI	BP	Hg-contaminated crushed light bulbs	5 gal	40	5,000 to 7,000	1.8	0.200 to 0.202	<0.00004 to 0.00005
DOE (1999a)	MI	BP	DOE Surrogate Wastes of nitrate salts and off-gas scrub solution	NR	58 to 70	1,400 to 1,900	1.7 to 2.0	540 to 650	<0.00004 to <0.00005
Wagh et al. (2000)[b]	MI	BP	DOE Ash ($HgCl_2$ at 0.5 wt%)	(100 g)	NR	NR	NR	40	<0.00085
Wagh et al. (2000)[b]	MI	BP	Delphi DETOX (with 0.5 wt% each $HgCl_2$, Ce_2O_3, $Pb(NO_3)_2$)	(100 g)	NR	NR	NR	138 to 189	<0.00002 to 0.01
Wagh et al. (2000)[b]	MI	BP	Soil ($HgCl_2$ at 0.5 wt%)	(100 g)	NR	NR	NR	2.27	<0.00015
Wagh and Jeong (2001)[c]	MI	BP	Detox Wastestream ($HgCl_2$ at 0.5 wt%)	(162 to 500 g)	60 to 78	NR	NR	250[a]	4.7 to 15.1
Wagh and Jeong (2001)[c]	MI	BP	Detox Wastestream (Hg at 0.5 wt%)	(162 to 500 g)	60 to 78	NR	NR	250[a]	7.19 to 7.64

(a) Untreated waste TCLP not reported, so estimated based on total Hg level in waste divided by 20.
(b) Potassium sulfide was added to reduce metal leachability.
(c) Sodium sulfide nonahydrate was added to reduce metal leachability.

Singh et al. (1998) demonstrated the macroencapsulation of four waste streams with CBPC including cyrofractured debris, lead bricks, lead-lined plastic gloves, and mercury-contaminated crushed light bulbs. The cyrofractured debris consisted of metals, wood, bricks, rocks, and plastics. Some material handling and size reduction (e.g., shredding) was needed to fit the wastes into the waste disposal drum. The study was a bench-scale project with waste form sizes ranging from 1.2 to 5 gallons. In general, debris fragments were sized to be less than one third the diameter of the drum. The CBPC fabrication process was approximately the same for each waste with the exception of minor formula changes in the wt% of water, ash, or binders and the addition of K_2S in the mixture for the mercury-contaminated crushed light bulbs.

Cryofractured Debris

For the cryofractured debris, the phosphate ceramic slurry was created with a premixed powder of calcined magnesium oxide and fly ash added to an acid phosphate solution in a Hobart mixer. The CBPC formula consisted of a ratio of 40 wt% ash, 40 wt% binder (MgO and KH_2PO_4 powders mixed in 1:1 molar ratio) and 20 wt% water. The slurry was mixed at low speed until it reached the desired consistency. The slurry then was poured into the drum containing the waste and was stirred continuously to assure homogeneity of the mixture. The temperature was monitored and peaked at approximately 72 ^oC (162 ^oF) and the CBPC set at around 55 ^oC (131 ^oF). The final waste forms had a waste loading of 35 wt% and a density of 1.81 g/cm^3.

Lead Brick and Lead-Lined Gloves

The low-level radioactive lead brick and lead-lined plastic gloves were encapsulated in CBPC formulated from 60 wt% ash, 25 wt% binder, and 15 wt% water. Macroencapsulation of the lead brick involved pouring a 2-inch lower base and allowing it to set for one hour until it could bear the weight of the lead brick. The macroencapsulation of the glove wastes was accomplished with the use of a plastic cage suspended in a 5-gallon pail.

Mercury Contaminated Light Bulbs

The mercury-contaminated crushed light bulbs were pre-treated by mixing with a potassium sulfide solution for approximately 1 hour. The glass was then set in CBPC with a similar formulation to the cryofractured debris. Mercury levels in the glass waste were around 200 parts per million (ppm). The crushed glass ranged in size from 2 to 3 cm long by 1 to 2 cm wide. During the mixing of the waste with the binder, the glass was crushed down to sizes less than 60 mm and a waste loading of approximately 40 wt% was achieved.

Each waste form was allowed to cure for about two weeks prior to performance testing. The density of the final waste forms was approximately 1.8 g/cm^3, the open porosity less than 4%, and the compression strengths between 5,000 and 7,000 psi. The cross sections of the final waste forms were observed to be very homogenous, dense, and free of air pockets. A complete, intact coating with continuous adhesion was observed around the lead brick and other wastes and no gaps were present at waste-binder interfaces. TCLP tests on the mercury-contaminated wastes showed 200 to 202 μg/L in the untreated wastes compared to <0.04 to 0.05 μg/L for the treated wastes. Key performance data from this study is summarized in Table 2-2.

A DOE study (1999a) was completed to test the effectiveness of CBPCs in the treatment of salt-containing, mercury-contaminated mixed wastes. A significant proportion of DOE mixed wastes contain greater than 15 wt% salts and these wastes are very difficult to treat with conventional methods. Salts adversely impact conventional cement matrices by causing a decrease in compressive strength and an increase in metal leachability. There is a demonstrated need to find encapsulation materials that can allow higher waste loadings to be achieved compared to conventional cement stabilization/solidification. The waste streams used in this study included saturated salt solutions ($NaNO_3$ and NaCl), activated carbon, ion exchange resins, spent incinerator off-gas scrub solution, and Na_2CO_3. These surrogate wastes were spiked with hazardous constituents including lead, chromium, mercury, cadmium, nickel, and trichloroethylene (TCE) at levels up to 1,000 ppm.

Waste loadings in CBPC of up to 70 wt% (40 wt% salt) were achieved during the study. Several performance tests were completed on the CBPC-encapsulated wastes, including compressive strength, U.S. EPA TCLP tests, and salt anion leaching tests per American National Standards Institute (ANSI) Method 16.1. The final CBPC waste forms fabricated with the saturated salt solutions had densities ranging from 1.72 to 1.8 g/cm^3 and compressive strengths ranging from 1,800 to 3,500 psi. The binder was amended with K_2S, which successfully stabilized mercury to meet the TCLP limit in these wastes. Anion leaching indexes of 6.9 and 6.7 were measured for chloride and nitrate, respectively, which barely passed the demonstration's criteria level (6.0). These results indicate that salt leaching may deteriorate the waste over time and that an additional binder or coating technique for the surface may be needed. Subsequently, some CBPC waste forms were coated in a commercial polymer to plug the surface pores and the combined leaching index of NO_3 and Cl was changed to 12.6, which indicated a reduction in leaching behavior. Key performance data from this study is summarized in Table 2-2.

Wagh et al. (2000) discusses the results of bench-scale studies for the encapsulation of mercury-contaminated surrogate wastes including DOE ash waste, secondary waste streams from the DETOX^{SM} wet oxidation process, and contaminated topsoil. The surrogate waste streams were dosed with mercuric chloride ($HgCl_2$) at 0.1 wt% to 0.5 wt% and also with other metals including lead and cesium. Initial tests showed that encapsulation with CBPC alone caused mercury leaching to decrease by a factor of three to five times. However, for adequate mercury stabilization, Wagh et al. determined that a small amount of Na_2S or K_2S should be used in the binder. For use with CBPC, the K_2S formulation was initially deemed to be the most appropriate because the CBPC binder is a potassium-based material. Other potential additives for mercury stabilization referenced by the author include H_2S or NaHS. In this study, K_2S was mixed directly with MgO and KH_2PO_4 powders to form one binder powder. The optimal range of K_2S in the binder powder was found to be 0.5 wt% and it was also established that levels significantly above this dose resulted in the formation of Hg_2SO_4, which has a much higher solubility than HgS (Hg_2SO_4 has a solubility product of 7.99×10^{-7} versus HgS with a solubility product of 2.0×10^{-49}). All of the surrogate wastes were successfully treated to levels below the U.S. EPA TCLP criteria for mercury from initial, untreated TCLP levels ranging from 2.27 mg/L in the soil to 189 mg/L for the iron phosphate wastes. Long-term (90-day) leaching tests were also performed on the waste forms. It was determined that the diffusion of mercury through the CBPC matrix is 10 orders of magnitude lower than in cement systems. Key performance data from this study is summarized in Table 2-2.

Wagh and Jeong (2001) continued work related to the encapsulation of DETOX^{SM} wastes. The study was concerned with the effect of haematite (Fe_2O_3) on the fabrication and setting of the CBPC waste form. The DETOX^{SM} wastes contained approximately 95 wt% Fe_2O_3, which was found to be highly reactive and caused the CBPC slurry to set too quickly before mercury could be effectively fixed into HgS. Additional tests were conducted in order to modify the CBPC fabrication process to account for the reactive nature of these wastes. Two surrogate wastes were created including a waste stream with 0.5 wt% $HgCl_2$ and 94.32 wt% Fe_2O_3 and a waste stream with 0.5 wt% HgO and 95 wt% Fe_2O_3. Two samples of each surrogate waste were pretreated with sodium sulfide nonahydrate ($Na_2S.9H_2O$) for two hours, which allowed sufficient time for the mercury to form HgS. The binder was then added and the slurry was mixed until it set. The CBPC samples were cured for three weeks and subjected to the U.S. EPA TCLP test. Final TCLP results for the treated $HgCl_2$ waste ranged from 4.7 to 15.1 μg/L and the HgO wastes ranged from 7.19 to 7.64 μg/L. Waste loadings ranged from 60 to 78 wt%. Setting times were rapid (10 to 18 minutes) and the authors suggested that it may be possible in large-scale systems to slow down the reaction by adding boric acid (at <1 wt%). Key performance data from this study is summarized in Table 2-2.

The following is a list of advantages and limitations associated with the use of CBPC for the encapsulation of hazardous wastes:

Advantages

- Waste stabilization is due to both chemical stabilization and physical encapsulation.
- Low temperature process (<80 ^oC or 176 ^oF).
- CBPC can be used to treat dry solids, sludges, and liquids.
- Unlike SPC, CBPC requires no additional heat input.
- High waste loading (up to 78 wt%) minimizes disposal volumes.
- Superior water tightness and chemical resistance compared to Portland cement.
- Simple to implement since mixing and pouring equipment is readily available.
- Nonflammable and stable and safe with oxidizing salts.
- No secondary wastes are generated.
- The process does not generate potentially hazardous off-gasses.

Limitations

- Pretreatment with K_2S or other compounds is needed for chemical stabilization of mercury; CBPC alone is not enough.
- Excess sulfide will increase the leachability of mercury, so careful processing is needed.
- Some waste constituents (e.g., haematite) may accelerate setting times and decrease workability of the CBPC slurry.
- Only limited data is available to support the long-term effectives and durability of CBPC waste forms.
- For high salt wastes, the leaching of salt anions over time could deteriorate the integrity of the waste. A polymer coating of the waste form may be needed to decrease the leaching of salt anions.

2.3 Polyethylene Encapsulation

Polyethylene is a thermoplastic material or a noncross-linked linear polymer that melts and liquefies at a specific transition temperature (120 ^{o}C or 248 ^{o}F). Polyethylene physically encapsulates the waste and does not interact with or chemically alter the waste materials. Polyethylene is readily available as a post-consumer recycled material (e.g., low-density polyethylene [LDPE] and high-density polyethylene [HDPE] used in commercial packaging/containers). It also has good chemical resistance and is water insoluble. According to Kalb et al. (1997) the physical properties of LDPE are better suited to encapsulation because HDPE requires greater temperatures and pressures during processing and mixing with wastes. LDPE with a high melt index from 50 to 55 g/10 minutes is reported to provide the optimal melt viscosity for mixing with wastes (Kalb et al. under U.S. Patent No. 5,649,323).

Figure 2-3 provides a simplified block diagram for the polyethylene macroencapsulation process. The key equipment used in this process typically includes a polymer extruder and feed hoppers. There are three types of extruder units including a single screw extruder, an intermeshing counter-rotating twin-screw extruder, and an intermeshing co-rotating twin-screw extruder. These extruders melt the polyethylene feed through both heat generated by friction from the rotating screw and supplemental barrel heaters. Single screw extruders are well-suited to both macroencapsulation and microencapsulation and have been used in the plastics industry for over 50 years (Kalb et al., 1992). Kinetic mixers have also been used for polyethylene encapsulation (Jackson, 2000). Polyethylene macroencapsulation typically involves the use of a basket placed inside a drum to allow at least a 1-inch barrier around the waste material. Molten polyethylene is then poured from an extruder over and around the waste in the drum. The drum can be rotated to ensure a more uniform distribution of the molten plastic. (An alternative to on-site pouring is the use of pre-manufactured containers.) Polyethylene microencapsulation typically involves directly mixing the waste material and polyethylene at an elevated temperature (typically 120 to 150°C or 248 to 302°F) in an extruder. The mixture of waste material and polyethylene is then poured into a drum and allowed to set. Microencapsulation may require several pretreatment steps, including drying of wet wastes and physical separation to resize or improve the particle distribution of the waste (Faucette, 1994). At the Envirocare facility in Utah, the optimal processing parameters for microencapsulation in a single screw extruder were determined to be a maximum of 2% moisture content and a 3-mm particle size limit (Jackson, 2000). In addition, off-gas treatment is needed for any water vapor, volatile organic compounds (VOCs), or volatile metals (e.g., arsenic and mercury) in the waste (Faucette, 1994). Polyethylene microencapsulation and macroencapsulation services are commercially available. In 1998, the Envirocare facility in Utah installed and permitted a single screw extruder system that can process up to 5 tons of waste per day. The final waste forms are typically set in 30- to 55-gallon drums and have a minimum exterior surface coating of LDPE of 1 to 2 inches (Jackson, 2000).

Several studies have been carried out using polyethylene for both macroencapsulation and microencapsulation of hazardous wastes, including Faucette et al. (1994), Burbank and Weingardt (1996), and Carter et al. (1995). In addition, several commercial vendors (e.g., Chemical Waste Management, Boh Environmental, and Ultra-Tech, International) provide macroencapsulation services with pre-manufactured HDPE containers. Encapsulation with polyethylene has been demonstrated with numerous waste streams including mixed waste salts,

sludges, and ash. Because macroencapsulation is BDAT for radioactive lead solids, several studies deal with macroencapsulation of lead brick and shielding waste materials including Faucette et al. (1994) and DOE (1998). One study was found which dealt with the encapsulation of radioactive, concentrated salts and basin sludges with low levels of mercury ranging from 1.3 to 9.2 ppm (Burbank and Weingardt, 1996). Carter et al. discusses the difficulties encountered with the microencapsulation of high-level arsenic wastes due to the high volatility of arsenic trioxide. In general, there is little performance data available on the effectiveness of polyethylene encapsulation of mercury-containing wastes. Key performance data from these studies is summarized in Table 2-3.

Faucette et al. used polyethylene for macroencapsulation and microencapsulation of a variety of mixed waste streams from the DOE Rocky Flats Plant in Colorado. The purpose of the macroencapsulation demonstration was to compare two containment methods including physical contact (e.g., on-site pouring of the waste form) versus pre-manufactured inserts. The objectives of the microencapsulation demonstration were to identify optimal processing equipment, test various additives to reduce the leachability of metals in surrogate wastes, and complete a treatability study for actual salt wastes. Key performance data from this study are included in Table 2-3.

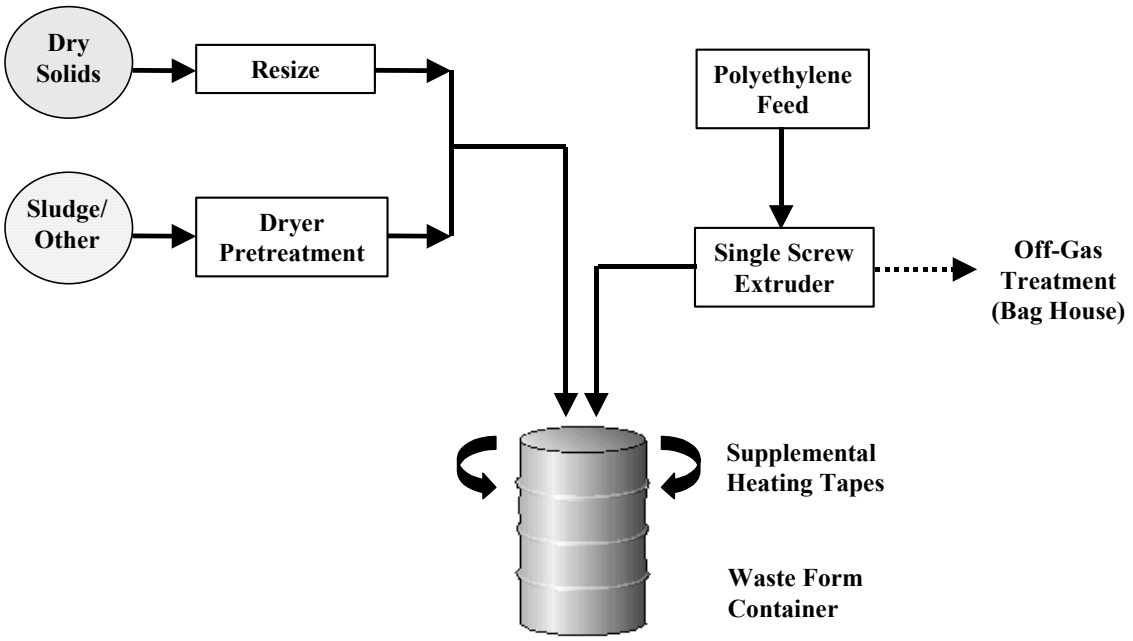

Figure 2-3. Polyethylene Macroencapsulation

Faucette et al. demonstrated the macroencapsulation of low-level, mixed wastestreams identified as combustibles (e.g., paper, cloth, plastics), laboratory glassware, scrap metals (e.g., pipe, valves, hand tools), and lead (e.g., sheet, bricks, tape). Two different approaches were used to create a 1-inch-thick polyethylene barrier around the wastes including physical contact and

Table 2-3. Key Performance Data for Polyethylene Encapsulation

Author/ Vendor	Type	Material	Scale	Waste Type	Waste Form Size	Waste Loading (wt%)	Compressive Strength (psi)	Density (g/cm^3)	Before Hg TCLP (mg/L)	After Hg TCLP (mg/L)
Faucette et al. (1994)	MA	LDPE	BP/F	Combustibles, laboratory glassware, scrap metals, and lead (e.g., sheet, bricks, tape).	5 to 10 gal	NR	NR	NR	NA	NA
Faucette et al. (1994)	MI	LDPE	BP/F	F006 Waste Code: Nitrate Salts with Cd, Cr, Pb, Ni, and Ag	NR	50	NR	NR	NA	NA
Burbank and Weingardt (1996)	MI	LDPE	BP	Ammonium sulfate/solar basin sludge	1.25 gal	40 to 50	1,088 to 2,465	NR	0.46 [9.2 ppm][(a)]	0.442 to 1.07
Burbank and Weingardt (1996)	MI	LDPE	BP	Solar basin sludge	1.25 gal	40 to 50	1,088 to 2,465	NR	0.065 [1.3 ppm][(a)]	0.107 to 0.122
Carter et al. (1995)	MI	HDPE	BP	As_2O_3	NR	(20 vol%)	NR	NR	NA	NA
Kalb et al. (1996)	MI	LDPE	BP	Off-gas scrub solution	NR	50 to 70	1,950 to 2,180	1.21 to 1.45	0.14	<0.009

(a) Untreated waste TCLP not reported, estimated by total Hg level in waste divided by 20.

pre-manufactured inserts. Polyethylene was used with a melt index of 50 to 200 grams/10 minutes per American Society for Testing and Materials (ASTM) D1238-90b. It was determined that LDPE experienced less cratering and cracking than HDPE and had a lower expansion coefficient (e.g., shrank less upon cooling). The basket holding the waste material in place also had to be flexible and yield as the polymer cooled and contracted. During scale-up to a 5-gallon container, Faucette et al. found that it was necessary to modify the process by using a "crock pot" to heat the waste form to control the temperature and viscosity of the polyethylene during the pour. Faucette et al. also stated that the basket required to hold the waste could cause potential leak paths. Macroencapsulation with a pre-manufactured polyethylene insert also was demonstrated.

The insert consisted of an open top, thick-walled polyethylene liner, which was placed into a 5-gallon metal container. The insert then was filled with waste and capped with molten polyethylene. The pre-manufactured insert resulted in a waste form of known thickness and only the cap needed to be poured on site.

One limitation of polyethylene encapsulation is that wastes must be dewatered prior to processing. Faucette et al. tested four different types of drying units for pretreatment of waste materials including a spray dryer, a horizontal thin film evaporator, a vertical thin film evaporator, and a horizontal rotary/blender dryer. The drying units were tested for their ability to concentrate a nitrate salt aqueous waste stream contaminated with various metals and high chlorides and sulfates. The horizontal thin film unit was chosen because it produced salts with the largest particle size and the highest bulk density. Several additives to the polyethylene were tested for their ability to reduce the leachability of cadmium, chromium, lead, nickel, and silver. The addition of surfactant (0.5 wt% sodium stearate) was found to improve the wetting of the salts by the polyethylene and reduce the leachability of cadmium and chromium. Calcium oxide and magnesium oxide also significantly reduced the TCLP results for cadmium and chromium. Carbon, alumina, diatomite, and class C fly ash were found to reduce chromium leachability by 93 to 98%, but cadmium was unaffected. It also was determined that excess water (e.g., >2 wt%) caused the salts to clump together, resulting in highly variable feed characteristics and a heterogeneous product.

Burbank and Weingardt explored the use of polyethylene for the microencapsulation of mixed wastestreams at the DOE site in Hanford, WA. Two wastes contained detectable levels of mercury along with other metals, including ammonium sulfate cake wastes with 9.2 ppm of mercury and solar evaporation basin sludge with 1.3 ppm of mercury. These wastes were incorporated into polyethylene at a 40 to 50 wt% loading. Prior to encapsulation, calcium oxide was added to the wastes to help reduce the leachability of metals. Based on TCLP results, the amendment of the wastes with calcium oxide did not reduce mercury leachability. Microencapsulation of the ammonium sulfate cake waste with polyethylene resulted in a mercury TCLP of 0.442 mg/L. With the addition of calcium oxide, this same wastestream had a mercury TCLP of 1.07 mg/L. It is clear from these results that, even at relatively low levels in waste, polyethylene encapsulation alone cannot adequately reduce the availability of mercury, and chemical stabilization (e.g., transformation to HgS) is necessary prior to the encapsulation of such wastes with polyethylene. Also, due to the high processing temperatures of polyethylene encapsulation, it is likely that a large fraction of mercury in these wastes will be volatilized

unless it has been chemically fixed. Key performance data from this study are included in Table 2-3.

Carter et al. used HDPE with a melting point of 130 ^{o}C (266 ^{o}F) and an operating temperature of 180 – 210 ^{o}C to microencapsulate powdered arsenic trioxide (As_2O_3). It was found that at a 20 volume percent (vol%) loading of this compound, the viscosity of the HDPE increased dramatically and the mixture became unworkable. Scanning electron microscope (SEM) micrographs showed that the arsenic trioxide had sublimed and recrystallized. When arsenic trioxide was stabilized to calcium oxide, the volatility decreased, but achievable waste loadings in HDPE remained low. Mercury and its compounds are also highly volatile compared to other metals (e.g., mercuric chloride sublimes at 300 ^{o}C or 572 ^{o}F), so the results of this study could provide some insight into the challenge of using polyethylene to process wastes containing high levels of arsenic and mercury. Key performance data from this study are summarized in Table 2-3.

There are several vendors that provide macroencapsulation services with pre-manufactured HDPE containers including Chemical Waste Management, Boh Environmental, and Ultra-Tech, International. These macroencapsulation methods are allowed under the alternative debris standards (40 CFR 268.45) because the definition of macroencapsulation for debris does not preclude the use of materials that meet the definition of tank or container (40 CFR 260.10). Chemical Waste Management provides ½-inch-thick HDPE vaults measuring 21 feet by 7 feet for the disposal of hazardous waste debris. A 3-inch-thick soil liner is used in the vault to provide a physical cushion between the bottom of the vault and the debris. Soil or sand is typically used to fill any void spaces around the debris. Once the vault is full, the lid is secured to the vault with adhesives and screws. The vault then is placed in a subtitle C landfill. Chemical Waste Management also provides 225-millimeter HDPE-lined roll-off boxes for hazardous waste debris disposal. Boh Environmental's Arrow-Pak™ technology consists of compacting 55-gallon drums filled with mixed/hazardous waste debris into 12-inch-thick pucks. The compacted drums are loaded into an 85-gallon metal overpack drum and then into a 1-inch-thick HDPE tube about 21 feet in length and 30 inches in diameter. Each tube fits the equivalent of 21 55-gallon drums. Both ORNL and DOE's Hanford have used this technology for the macroencapsulation of mixed waste debris. This technology achieves a mixed waste debris volume that is typically one-fourth that of on-site macroencapsulation with polyethylene (INEL, 2002). Ultra-Tech, International offers a series of pre-manufactured, medium-density polyethylene containers for macroencapsulation. The containers can be custom-made in any size, but have been manufactured to over-pack one 55-gallon drum to containers 52 inches in diameter and 20 feet in length. A resistance wire system is embedded in the lid of each container. Once the debris waste is in place, an electrical current is applied to the wires, heating them up to melt the polyethylene, and creating an effective seal around the top of the container. This technology is currently being tested by DOE's Mixed Waste Focus Area Program (Ultra-Tech, International, 2002).

The following is a list of advantages and limitations associated with the use of polyethylene for the encapsulation of hazardous wastes:

Advantages

- Polyethylene has a high mechanical strength, flexibility, and chemical resistance.
- Polyethylene is highly resistant to biological degradation.
- Polyethylene allows higher waste loadings (up to 70 wt%) compared to conventional Portland cement.
- Polyethylene is readily available in postconsumer recycled forms.
- Equipment is commercially available and the process can be automated, so the operator input requires only drum placement.
- Pre-manufactured vaults and containers can be used, which provide a final waste form of known barrier thickness and integrity.
- Because HDPE is used in landfill liners, extensive studies have been performed to document the chemical resistance and long-term durability of HDPE.

Limitations

- External heating is required and the process occurs at a higher temperature than the SPC and CBPC methods.
- Polyethylene does not chemically incorporate the waste, and with mercury-containing wastes volatilization may be a significant concern.
- Chemical stabilization of mercury-contaminated wastes prior to encapsulation may be necessary to meet TCLP requirements.
- The encapsulation of high-level arsenic wastes with polyethylene is problematic due to the sublimation of arsenic compounds at high temperature (>200 ^{o}C).
- Small quantities of secondary waste are generated.
- For large-scale or on-site pouring, LDPE is preferred because HDPE is prone to cratering, cracking, and excessive shrinking.
- LDPE is intolerant of the presence of free liquids and organics.
- Wastes must be pretreated to remove moisture.
- Molten polyethylene can cause severe burns, so extra safety precautions are necessary.
- Small quantities of secondary waste are generated.

2.4 Other Encapsulation Materials

Several other materials have been developed and demonstrated for the encapsulation of mercury-containing hazardous wastes including asphalt, polyester and epoxy resins, synthetic elastomers, polysiloxane, sol-gels (e.g., polycerams), and Dolocrete^{TM}. Key performance data are summarized in Table 2-4. In addition, a variety of materials currently available for the encapsulation of other metal-containing wastes are discussed.

2.4.1 Asphalt

Asphalt or bitumen has been used to microencapsulate soil contaminated with low-levels of heavy metals (Smith et al., 1995 and Hubbard et al., 1990). Radian corporation reported using cold-mix asphalt to microencapsulate soil contaminated with mercury (78 mg/kg). The material had a compressive strength of 176 psi. Hot-mix asphalt was deemed to be inappropriate because the elevated temperatures could promote the volatilization of mercury (SAIC, 1998). Kalb et al.

(1996) discusses the microencapsulation of up to 60 wt% of a mixed waste incinerator off-gas scrub solution with asphalt. The mercury TCLP in the untreated wastes was 0.14 mg/L versus <0.009 mg/L in the asphalt microencapsulated waste. Compressive strengths averaged 570 psi in the final waste forms.

2.4.2 Polyester and Epoxy Resins

Polyester is an example of a thermosetting resin or a cross-linked polymer that undergoes a chemical reaction to solidify. Several thermosetting resins have been tested for the encapsulation of salt-containing mixed wastes including orthophthalic polyester, isophthalic polyester, vinyl ester, and a water-extendible polyester. These wastes contained metals, including mercury, at the 1,000 ppm level. With polyester resins, waste loadings of 50 wt% were achieved for unconcentrated spent off-gas scrub solutions and 70 wt% for nitrate/chloride salts. In addition, compressive strengths ranged from 5,100 to 6,200 psi. Mixed waste, salt surrogate TCLP tests for mercury ranged from <0.01 to 0.2 mg/L (DOE, 1999b). Orebaugh (1993) reported using several epoxy resins (e.g., Stycast 2651 and Thermoset 300) to macroencapsulate mixed waste, lead billets. The waste forms were subjected to 6-foot drop tests to gauge their stability and mechanical strength.

2.4.3 Synthetic Elastomers

Synthetic elastomers are materials having properties similar to natural rubber and have been used in the microencapsulation and stabilization of metal-contaminated wastes. Carter et al. explored the use of styrene-butadiene rubber (Solprene 1204) for the encapsulation of powdered arsenic trioxide (As_2O_3). Up to 64 wt% of arsenic trioxide was incorporated into the rubber, but beyond this level the rubber became unworkable. Meng et al. (1998) reports using tire rubber for the immobilization of mercury-contaminated soils. A clay-loam soil was spiked with mercuric oxide and mercuric chloride at 300 mg/kg. Acetic acid leachate tests showed a reduction from 3.5 mg/L in the untreated soil to 0.034 mg/L in the soil mixed with tire rubber. The used tire rubber contained approximately 2 to 4% sulfur and less than 32% carbon black.

2.4.4 Polysiloxane

Polysiloxane or ceramic silicon foam (CSF) consists of 50 wt% vinyl-polydimethyl-siloxane, 20 wt% quartz, 25 wt% proprietary ingredients, and less than 5 wt% water. The use of this material for encapsulation is patented by Orbit Technologies. The material sets at room temperatures (30 ^{o}C or 86 ^{o}F) and is resistant to extreme temperatures, pressures, and chemical exposure. The polysiloxane technology was demonstrated on salt waste surrogates, which were spiked with lead, mercury, cadmium, and chromium at 1,000 ppm levels. Up to 50 wt% waste loading was demonstrated. The final waste form had a compressive strength of 600 psi at 40 wt% loading. For high chloride salt wastes, the mercury TCLP was 0.01 mg/L and for high nitrate salt wastes the mercury TCLP was 0.06 mg/L. The final waste forms for both waste types did not pass for chromium. The authors recommend pretreatment for the chemical stabilization of wastes with metals at levels greater than 500 ppm (DOE, 1999c). In addition, Miller et al. (2000) reports on the use of silicone foam to encapsulate a DOE surrogate waste containing high levels of chromium. Salt waste loadings of up to 48 wt% were achieved in this study.

Table 2-4. Key Performance Data for Various Encapsulation Materials

Author/ Vendor	Type	Material	Scale	Waste Type	Waste Form Size	Waste Loading (wt%)	Compressive Strength (psi)	Density (g/cm^3)	Before Hg TCLP (mg/L)	After Hg TCLP (mg/L)
Kalb et al. (1996)	MI	Asphalt	BP	Off-gas scrub solution	NR	30 to 60	540-610	1.08 to 1.42	0.14	<0.009
Radian[(b)]	MI	Asphalt	F	Soil (Hg 78 mg/kg)	NA	NR	176	NR	NR	NR
DOE (1999b)	MI	Polyester	BP	Salt-containing mixed wastes	NR	50	5,100 to 6,200	NR	50[(a)]	<0.01 to 0.2
Orebaugh (1993)	MA	Epoxy	BP	Mixed waste, lead billets	5 gal	NR	NR	1.43 to 1.5 (Resin Only)	NA	NA
Carter et al. (1995)	MI	Styrene-butadiene rubber	BP	As_2O_3	NR	64	NR	1.7 (Rubber Only)	NA	NA
Meng et al. (1998)	MI	Tire Rubber	BP	Soil (Hg 300 mg/kg)	(100 g)	(4 g rubber /100 g soil)	NR	NR	(3.5 acetic acid)	(0.034 acetic acid)
DOE (1999c)	MI	Poly-siloxane	BP	Salt-containing mixed wastes	NR	50	420 to 637	NR	50[(a)]	0.01 to 0.06
DOE (1999d)	MI	Sol-Gels	BP	Salt-containing mixed wastes	NR	60 to 70	1,050 to 1,513	NR	50[(a)]	0.044 to 0.23
Dolomatrix (2001)	MI	Dolocrete™	F	Hg-waste at 15,300 mg/kg	NR	NR	145	NR	765[(a)]	<0.1

(a) Untreated waste TCLP not reported, estimated by total Hg level in waste divided by 20.
(b) SAIC (1998).

2.4.5 Sol-Gels

Sol-gels or polycerams are a hybrid material derived from the chemical combination of organic polymers and inorganic ceramics. A DOE study (DOE, 1999d) explored the use of a polyceram consisting of a polybutadiene-based polymer combined with silicon dioxide for the stabilization of high salt wastes. The salt waste surrogates contained lead, chromium, mercury, cadmium, and nickel at 1,000 ppm levels. The polymer and silicon dioxide are combined first and then mixed with the waste and then solidified to encapsulate the waste. The setting of the waste form takes place at temperatures ranging from 66 to 70 ^{o}C (151 to 158 ^{o}F). Waste loadings from 30 to 70 wt% were demonstrated. Compressive strengths of the final waste forms ranged from 137 to 1,513 psi. The initial waste forms in the demonstration had a high open porosity and did not pass the TCLP test for mercury. Another set of waste forms were fabricated and subjected to a secondary infiltration of polyceram solution after initial drying. The second set of tests was able to demonstrate a decrease in the mercury TCLP to 0.044 mg/L.

2.4.6 Dolocrete™

Dolocrete™ is a proprietary calcined dolomitic binder material that can be used for the microencapsulation of inorganic, organic, and low-level radioactive waste. Dolocrete™ is reported to successfully encapsulate wastes containing aluminum, antimony, arsenic, bismuth, cadmium, chromium, copper, iron, lead, mercury, nickel, tin, and zinc. The encapsulation of mining waste with up to 590,000 mg/kg of arsenic resulted in a TCLP of 3.9 mg/L, which meets the current arsenic TCLP limit of 5 mg/L. Mercury-contaminated wastes with up to 15,200 mg/kg were treated to reach a TCLP level of <0.1 mg/L. Compressive strengths of the final waste forms often exceed 145 psi (Dolomatrix, 2001).

2.4.7 Materials Used With Other Metals

For the stabilization/solidification of other hazardous metal wastes, cement, Pozzolan, and lime are the most commonly used encapsulation materials; however, research continues into the use of other binders and additives to enhance performance of the final waste form and to reduce project costs (Conner and Hoeffner, 1998). Figure 2-4 lists binders and additives that have been found to decrease metal leachability including fly ash, clays, slags, iron compounds, activated carbon, and other materials. In addition to conventional cement, Pozzolan, and lime-based binders, another encapsulation material reported in the literature is proprietary silicate binders. Several vendors and authors report using silicate-based materials for the encapsulation of metal-contaminated wastes including Chemfix Technologies, Inc.; Silicate Technology Co. (STC), Mitchell et al. (2001); and Evangelou (2000).

Other

•Type I, II, V Portland cements
•Class F fly ash
•Cement kiln dust
•Lime kiln dust
•Slag
•Sodium silicate and proprietary polysilicate mixtures
•Dolocrete™
•Calcium carbonate (limestone)
•Calcium sulfate (gypsum)
•Iron oxide (hematite)
•Calcium phosphate (apatite)
•Organophillic clay with additives

pH Control

•Sulfuric acid
•Phosphoric acid
•Buffer solution

Other Pretreatment Additives

•Potassium permanganate (oxidation)
•Hydrogen peroxide (oxidation)
•Calcium hypochlorite (oxidation)
•Potassium or sodium persulfate (oxidation)
•Ferric or calcium chloride (precipitation)
•Ferric or ferrous sulfate (precipitation)
•Magnesium oxide (adsorbent)
•Activated carbon (adsorbent)

Note: Based on information from Wickramanayake et al., 2001; Conner and Hoeffner, 1998; Sun et al., 2001; Dutre and Vandecasteele, 1995; and Rha et al., 2000.

Figure 2-4. Materials and Additives for Stabilization/Solidification of Other Metals

Chemfix Technologies, Inc., has developed a stabilization/solidification process using proprietary additives of soluble silicates and calcium-containing reagents. This process was tested under the U.S. EPA's Superfund Innovative Technology Evaluation (SITE) program in March of 1989. The Chemfix™ process was most successful in reducing the leachability of cadmium, copper, chromium, lead, nickel, and zinc. However, some difficulty was experienced in the treatment of arsenic and mercury-containing wastes. Before treatment, lead TCLP levels ranged from 390 to 890 mg/L in contaminated soil. After treatment, lead TCLP levels ranged from <0.5 to 47.0 mg/L or a 94% to >99% reduction in lead leachability. Initial copper TCLP

levels in contaminated soil ranged from 12 to 120 mg/L, whereas after treatment levels were reduced to 0.54 to 0.60 mg/L or a 96% to >99% reduction. The compressive strength of the final waste form was 90 psi (U.S. EPA, 1991).

STC developed a stabilization/solidification process, which relies upon the use of a proprietary silicate-mineral reagent that binds the metals into a layered alumino-silicate structure. The STC process was tested in the U.S. EPA SITE program in November of 1990. The SITE program involved the testing of the STC process on soils contaminated with both inorganic constituents (e.g., arsenic, chromium, and copper) and organic constituents (e.g., pentachlorophenol). Before treatment, arsenic TCLP levels ranged from 1.1 to 3.3 mg/L. After treatment, arsenic TCLP levels ranged from 0.09 to 0.88 mg/L or a 35% to 92% reduction. Chromium TCLP levels actually increased as a result of treatment from <0.05 to 0.27 mg/L before treatment to 0.19 to 0.32 mg/L after treatment. Initial copper TCLP levels ranged from 1.4 to 9.4 mg/L and were reduced by 90% to 99% to 0.06 to 0.10 mg/L. The compressive strength of the final waste forms ranged from 760 to 1,400 psi (U.S. EPA, 1992).

Mitchell et al. reports using silica microencapsulation for the treatment of aqueous acid rock drainage (ARD), which contained elevated levels of aluminum, arsenic, copper, iron, nickel, and sulfate. The process employs a proprietary mixture of chemicals referred to as KB-1™, which includes a silica-based reagent to chemisorb the metals from the aqueous phase into a solid matrix. The sludge generated from this water treatment process was able to meet TCLP limits, which eliminated the need to dispose of the material as a hazardous waste. Evangelou (2000) also discusses the treatment of ARD through the microencapsulation of pyrite with iron phospate or silicate binders. Silicate materials were found to reduce the leaching of sulfate from pyrite wastes relative to the control treatments with limestone and phosphate.

3.0 Cost and Vendor Information

Table 3-1 includes a summary of typical cost data, along with vendor information, for several materials used for the macroencapsulation/microencapsulation of hazardous wastes. Table 3-1 also includes typical costs for competing technologies for mercury-contaminated hazardous wastes including thermal recovery, acid leaching, and vitrification. However, it should be noted that both thermal recovery and acid leaching will generate highly concentrated, secondary waste streams that will ultimately have to be immobilized prior to disposal. In general, vitrification is best suited to low volatility metals as opposed to mercury and arsenic. The cost data presented in Table 3-1 are meant to provide an order-of-magnitude cost range for each technology. True technology costs will be specific to the waste type, waste chemical and physical properties, and the levels of contaminants in the waste.

Table 3-1. Summary of Cost and Vendor Information for Encapsulation and Other Treatment Technologies

Technology	Developer/Vendor	Estimated Full-Scale Costs	Reference
SPSS	Brookhaven National Laboratory, NY	$2.88/kg or $1.31 per lbs	Morris et al. (2002)
CBPC	Argonne National Laboratory, IL	$15.45 per kg or $7.00 per lbs	DOE (1999a)
Polyethylene (on-site pour)	Envirocare, UT	$90 to $100 per ft^3	DOE (1998)
Arrow-Pak (HDPE)	Boh Environmental, New Orleans, LA	880 drums for $1,100,000	(Hanford, 2002)
Ultra-Macroencapsulation System	Ultra-Tech, International, FL	30" dia, 40" height $480 to $700 6' x 6' x 20' $20,000	(Ultra-Tech, 2002)
Polyester Resin	SGN Eurisys Services Co., Richland, WA	$11.52 per kg or $5.22 per lbs	(DOE, 1999b)
Synthetic Elastomer	No vendor information available	$25 per ton of used tire rubber, 4 wt% in treated soil	Meng et al. (1998)
Polysiloxane	Orbit Technologies, Carlsbad, CA	$1,900 per ft^3	(DOE, 1999c)
Sol-Gels	Pacific Northwest National Laboratory, Richland, WA	NA	(DOE, 1999d)
Other Technologies			
Acid Extraction	Environmental Technologies International, Wyomissing, PA	$100 to $250 per ton	Mulligan et al. (2001)
Cement-Based Stabilization/Solidification	Various	$16.37 per kg or $7.42 per lbs $5,000 per ft^3	DOE (1999a) DOE (1999c)
DeHg®	Nuclear Fuel Services, TN	$8.48 per kg or $3.85 per lbs	Morris et al. (2002)
Thermal Recovery	Mercury Recovery Services, New Brighton, PA	$650 to $1,000 per ton	Mulligan et al. (2001)
X-Trax™ Thermal Desorption	Remediation Technologies, Tuscon, AZ	$100 to $600 per ton	Mulligan et al. (2001)
Vitrification	Westinghouse Science and Technology Center, Pittsburgh, PA	$400 to $870 per ton	Mulligan et al. (2001), (U.S. EPA, 1997)

Note: The cost data presented above are meant to provide an order-of-magnitude cost range for each technology. True technology costs will be specific to the waste type, waste chemical and physical properties, and the levels of contaminants in the waste.

4.0 Future Development and Research Needs

A large body of literature exists regarding the research and development of alternative materials to conventional Portland cement for the encapsulation of hazardous metal-containing wastes. SPC, CBPC, and polyethylene are the most established materials, and each has its advantages and disadvantages for use in the macroencapsulation or microencapsulation of mercury-containing hazardous wastes.

Although several studies were noted which demonstrated the successful encapsulation of high-level, mercury-containing wastes with SPC and CBPC, the body of evidence for competent polyethylene encapsulation is limited. The higher temperatures of the polyethylene process may pose some difficulty in effective encapsulation of these wastes due to the volatile nature of mercury compounds. A better understanding of the long-term stability of final waste forms may be needed for some binder materials. In general, the long-term stability of materials encapsulated with SPC or CBPC have not been addressed, except for encapsulated mixed wastes, which are extensively tested under NRC protocols. Improving the understanding of the kinetics of low-temperature processes such as SPC or CBPC could help in scale-up and process optimization. Also, a better understanding is needed regarding the role of excess sulfides in increasing mercury leachability. In addition, the performance objectives or acceptance criteria for macroencapsulated wastes could be standardized to provide guidance regarding the minimum layer thickness of the barrier, the expected long-term leaching performance of the final waste form, the target compressive strength, and the tolerance for void spaces in the final waste form.

Currently, few full-scale commercial applications of encapsulation technologies are available; however, further commercialization and technology transfer may occur if the demand for macroencapsulation increases as a result of changes in regulatory requirements. Both SPC and CBPC processes have been patented, but licensing of the technologies has generally been limited to one or two companies and application of these processes at the industrial-scale is limited. The Envirocare facility in Utah does have a full-scale system in place for polyethylene encapsulation. In addition, the use of pre-manufactured HDPE containers for macroencapsulation, as allowed under the U.S. EPA alternative debris standards, appears to offer a cost-effective solution to the disposal of hazardous waste debris. The use of other materials such as synthetic elastomers, polyester resins, polysiloxane, or sol-gels appears somewhat promising, but relatively few studies have been completed to date. With several of these materials, including polysiloxane and sol-gels, it appears that an additional chemical stabilization step may be needed when elevated levels of metals are present, since the TCLP criteria for mercury and chromium were not met in initial trails. In addition, the use of asphalt for encapsulation is most likely limited to contaminated soils with only low levels of mercury or other metals.

Because of the varied nature of industrial wastes, site-specific treatability tests will most likely be required for the selection of the most appropriate encapsulation material. The selection criteria should include chemical compatibility of the waste and binder materials, final waste form performance, technology implementability (e.g., the availability of processing equipment and vendor experience), safety and health issues, and project-specific estimated costs.

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