Advances in encapsulation technologies for the management of mercury-contaminated hazardous wastes ☆

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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 historic operations that have led to significant contamination and ongoing hazardous waste generation. This study was performed to evaluate whether the U.S. EPA could propose treatment and disposal alternatives to the current land disposal restriction (LDR) treatment standards for mercury. The focus of this article is on the current state of encapsulation technologies that can be used to immobilize elemental mercury, mercury-contaminated debris, and other mercury-contaminated wastes, soils, sediments, or sludges. The range of encapsulation materials used in bench-scale, pilot-scale, and full-scale applications for mercury-contaminated wastes are summarized. Several studies have been completed regarding the application of sulfur polymer stabilization/solidification, chemically bonded phosphate ceramic encapsulation, and polyethylene encapsulation. Other materials reported in the literature as under development for encapsulation use include asphalt, polyester resins, synthetic elastomers, polysiloxane, sol–gels, Dolocrete[TM], and carbon/cement mixtures. The primary objective of these encapsulation methods is to physically immobilize the wastes to prevent contact with leaching agents such as water. However, when used for mercury-contaminated wastes, several of these methods require a pretreatment or stabilization step to chemically fix mercury into a highly insoluble form prior to encapsulation. Performance data is summarized from the testing and evaluation of various encapsulated, mercury-contaminated wastes. Future technology development and research needs are also discussed.
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1. Introduction

The development of effective treatment options for mercury-contaminated wastes is a significant technical and practical challenge. There is little to no economic benefit derived from mercury recovery and recycling. In addition, effective treatment is often challenging due to the high toxicity, volatility, and environmental mobility of mercury and the varied nature and composition of industrial waste products. Principal industrial sources of mercury-contaminated wastes include chlor-alkali manufacturing, weapons production, copper and zinc smelting, gold mining, paint applications, and other processes [1]. Although industrial and com-

mercial 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 historic operations that have led to significant contamination and ongoing hazardous waste generation. This article focuses on the current state of encapsulation materials being used to immobilize elemental mercury, mercury-contaminated debris, and mercury-contaminated wastes, soils, sediments, and sludges. A technology overview is provided, along with a summary of the regulatory drivers and testing and evaluation criteria involved with implementation of these treatment approaches. The various encapsulation materials under investigation are discussed and future technology development and research needs are provided.

2. Technology overview

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. Encapsulation technologies are based primarily on solidification processes that act to "substantially reduce surface exposure to potential leaching media" (40 Code of Federal Register [CFR] 268.42). Encapsulation technologies can also involve a combination of physical entrapment through solidification and chemical stabilization through precipitation, adsorption, or other interactions. This combined treatment approach is sometimes referred to as stabilization/solidification.

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. For example, Singh et al. (1998) demonstrated the microencapsulation of mercury-contaminated crushed light bulbs in which the glass was crushed and mixed with the encasing material prior to solidification [2]. Mattus (1998) reported the macroencapsulation of mercury-contaminated lead pipes by pouring the encasing material over and around the lead pipes [3].

There are a wide variety of materials currently being evaluated in the scientific community and in industry for the encapsulation of hazardous wastes. This review focuses on the performance data related to the encapsulation of mercury-contaminated wastes, which has proven to be especially challenging given mercury's chemical and physical properties and the varied nature of industrial wastes. Conventional stabilization/solidification methods typically include the fixation of metals using Portland cement and fly ash. This produces an impermeable, solid waste form at a high pH (typically <10) 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. Arsenic and hexavalent chromium are also difficult to stabilize with cement-based processes due to the formation of anionic species that are soluble at high pH [4,5]. Therefore, a significant amount of research has gone into the development of other encapsulation materials that can be used as alternatives to the cement-based process. 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. Each encapsulation material identified in the literature 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.

3. Regulatory background

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 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 [4]. 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 [6]. This review provides details on the variety of encapsulation technologies available, but does not focus on other mercury treatment approaches (e.g. acid leaching, thermal recovery, etc.).

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 4–8 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. At a low pH, hydrogen sulfide gas may escape from the waste [7,8]. 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.

4. Testing and evaluation background

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 focus during macroencapsulation is to create an inert surface coating or jacket around the waste that substantially reduces the potential for exposure to leaching media [3]. For microencapsulated waste, the toxicity characteristic leaching procedure (TCLP) in the EPA publication SW-846, plays an important role in determining whether or not the material can be accepted by a landfill. According to the 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 generally must be treated to meet the Universal Treatment Standard (UTS) of 0.025 mg/L or less prior to disposal in a landfill. In addition, some states may set criteria that define hazardous wastes given the total metal concentration such as California's Total Threshold Limit Concentration (TTLC) of 20 mg/kg for mercury. The U.S. Nuclear Regulatory Commission (NRC) has also developed its own waste form acceptance criteria for mixed (e.g. radioactive) wastes. In general, NRC waste form testing procedures examine the influence of various environmental factors including the effect of thermal cycling and immersion on compressive strength, the impact of biodegradation and irradiation on waste form stability, and the long-term leaching behavior.

5. Encapsulation materials review

The following materials were identified during a comprehensive literature review as having been used for the encapsulation of mercury-contaminated wastes.

6. Sulfur polymer stabilization/solidification

The SPSS process can be used to 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 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 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 [9].
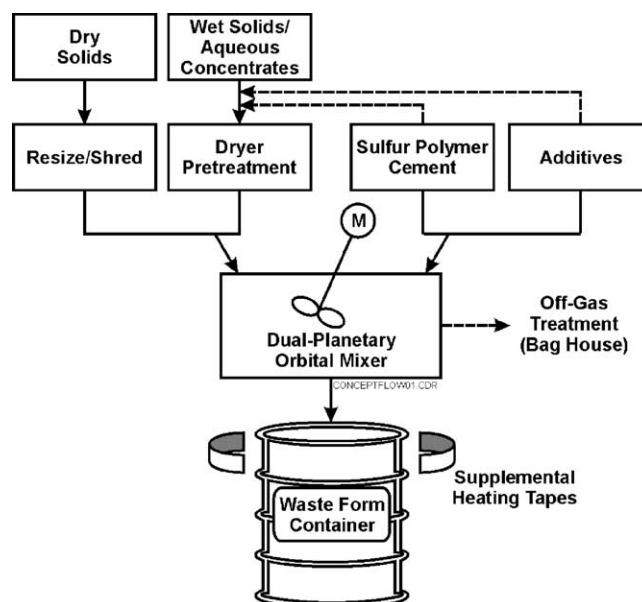

Fig. 1. Sulfur polymer stabilization/solidification process.

Fig. 1 provides a simplified block-diagram for the SPSS encapsulation process [10]. For macroencapsulation, molten SPC is poured over and around the waste or debris and is then allowed to set into a monolithic waste form. The recommended mixing temperature for SPC is between 260 and 280 °F. For microencapsulation of liquid, elemental mercury, a two-stage process is followed that 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 104 °F 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. An inert gas atmosphere is also used in the vessel to prevent the formation of mercuric oxide. Next, additional SPC is added and the mixture is heated to 266 °F to form a homogenous molten liquid. The liquid 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. BNL recently 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 [11,12].

Several studies have been completed regarding the use of SPC for heavy metal-contaminated wastes [13,3,14,9]. Key performance data from these studies are provided in Table 1.

Fuhrmann et al. (2002) presents the results from a bench-scale SPSS treatment of radioactive elemental mercury. The study explored three issues including the leachability of the treated waste, the formation of mercuric sulfide, and mercury vaporization during processing. Microencapsulation of the

Table 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)	Hg leachate untreated (mg/L)	Hg leachate treated (mg/L)
Mattus (1998)	MA	BP	Mixed waste cadmium sheets	5 gallons	15.8–28.6	NR	NR	NA	NA
Mattus (1998)	MA	BP	Mixed waste lead pipes/gloves contaminated with Hg	5 gallons	31.3–38.8	NR	NR	NA	NA
Fuhrmann et al. (2002)	MI	BP	Radioactive Hg^0	5 gallons	33.3	NR	NR	2.64	0.020 to >0.40
Fuhrmann et al. (2002)	MI	BP	Radioactive Hg^0 with 3 wt.% triisobutyl phosphine sulfide additive to SPC	5 gallons	33.3	NR	NR	2.64	>0.40
Fuhrmann et al. (2002)	MI	BP	Radioactive Hg^0 with 3 wt.% Na$_2$S·9H$_2$O additive to SPC	5 gallons	33.3	NR	NR	2.64	0.0013–0.050
Darnell (1996)[a]	MI	BP/F	Metal oxides including Hg, Pb, Ag, As, Ba, and Cr at 5 wt.% each	NR	40	4000	NR	250[b]	<0.2
Kalb et al. (1996)	MI	BP	Mixed waste off-gas scrub solution	NR	25–45	3850–8160	1.86–1.94	0.14	<0.009
Pildysh Technologies, Inc.	MI	BP	Mine tailings	<100 μm	~40%	NR	NR	413	<0.1

BP: bench-scale/pilot-scale; F: full-scale; MA: macroencapsulation; MI: microencapsulation; NA: not applicable; NR: not reported; TCLP: toxicity characteristic leaching procedure.

[a] Sodium sulfide nonahydrate was added to reduce metal leachability.

[b] Untreated waste TCLP not reported, so estimated based on total Hg level in waste divided by 20.

elemental mercury with SPC alone resulted in TCLPs ranging from 20 to >400 μg/L. Treatment using a 3 wt.% sodium sulfide nonahydrate additive resulted in TCLPs ranging from 1.3 to 50 μg/L. The authors used x-ray diffraction studies to determine that elemental mercury and SPC reacted to form primarily the more soluble, meta-cinnabar form of mercuric sulfide. However, elemental mercury and sodium sulfide nonahydrate formed primarily cinnabar, which explains the improved leaching behavior in those tests. The results of further studies also demonstrated that mercury volatilization was reduced through the treatment with sodium sulfide. Headspace measurements for elemental mercury alone 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 [13].

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 [3,15]. 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: (i) 457 lb of cadmium sheets and (ii) 448 lb

of lead pipes contaminated with mercury. It was found that preheating the debris to 284–302 °F for 6 h helped to prevent fast cooling of the SPC at the waste–binder interface during the pour. Preheating also helped to reduce the formation of air pockets. Vibrating the container during and after the SPC pour also improved setting of the waste form. Heating tapes were used to maintain a target temperature of 320 °F at the top portion of the container. This allowed air bubbles from the setting to escape. The optimal additional heating time was determined to be 10 h after the pour had ended. During testing, examination of the waste form cross sections revealed good contact between the debris pieces and SPC and no identifiable interface between pour layers. No H$_2$S or SO$_2$ off-gasses were detected during the tests. The investigators were able to incorporate up to 28.6 wt.% of the cadmium sheets and up to 38.8 wt.% of the mercury-contaminated lead pipes in the final waste forms [3,15].

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 wastes including dehydrated boric acid salts, incinerator hearth ash, mixed waste fly ash, and dehydrated sodium sulfate salts. 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$_2$S·9H$_2$O) was added to the SPC mixture to con-

vert metal oxides to more leach-resistant metal sulfides. The U.S. EPA TCLP limits were achieved for all metals [9].

Based on the information reviewed from the sources previously identified, some of the advantages of the SPSS process are as follows: (i) high concentration mercury wastes can be effectively treated, including elemental mercury; (ii) relatively low temperature process (260–280 °F); (iii) superior water tightness (e.g., low permeability and porosity) compared to Portland cement; (iv) high resistance to corrosive environments (e.g., acids and salts); (v) high mechanical strength; (vi) simple to implement because mixing and pouring equipment is readily available; (vii) easier to use than other thermoplastics, like polyethylene, because of its low viscosity and low-melt temperature; (viii) SPC can be remelted and reformulated.

Pildysh Technologies, Inc. has developed a proprietary, thermoplastic sulfur-based technology, called TerraBond[TM], for encapsulating and stabilizing hazardous wastes. The primary physical encapsulation is induced by allotropic sulfur crystal conversions. A hydrophobic sealant (secondary encapsulation) applied to the pellet surface provides a barrier against contaminant leaching.

One must also be aware of the limitations of the above-mentioned processes. Some of them are as follows: (i) volatile losses of mercury may occur and engineering controls are needed; (ii) aqueous wastes must be dewatered prior to processing; (iii) SPC can develop an excess of voids or air pockets if cooled too quickly; (iv) metal debris or pieces with large thermal mass may require preheating to prevent the formation of air pockets; (v) not compatible with strong alkaline solutions (>10%), strong oxidizing agents, aromatic or chlorinated solvents, or expanding clays; (vi) SPC handling requires the use of engineering controls to mitigate possible ignition and explosion hazards; (vii) if excessive temperatures are created, SPC will emit hydrogen sulfide gas and sulfur vapor.

7. 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. Similar to SPSS, 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} [7]. CBPCs are formed 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 potas-
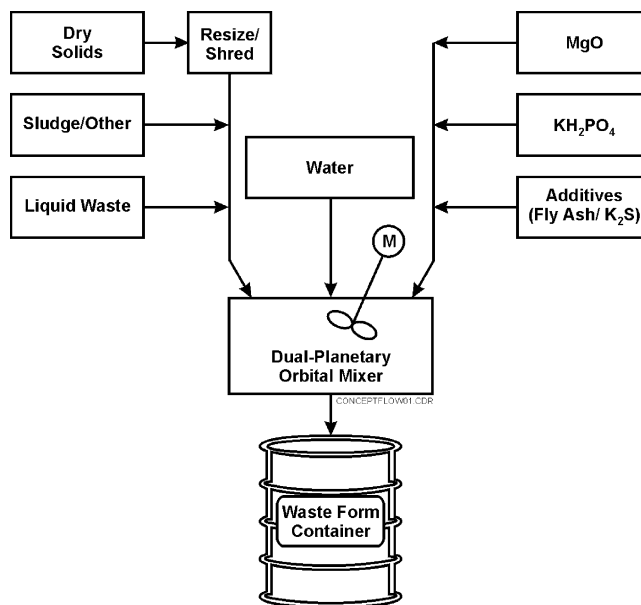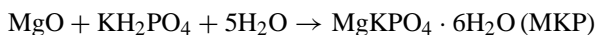

Fig. 2. Chemically bonded phosphate ceramic process.

sium phosphate hydrate as shown in the reaction below:

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

Iron oxide phosphates can also be used to form a low-temperature ceramic, but research into the use of this material is limited [16].

Fig. 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 the stoichiometric water content. Next, calcined magnesium oxide and monopotassium phosphate binders are ground to a powder and blended in a one-to-one molar 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 min. Under most conditions, heat from the reaction causes the waste matrix to reach a maximum temperature of approximately 176 °F. After mixing is stopped, the waste form typically sets in about 2 h and cures in about 2 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. 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 [17].

Several detailed studies have been completed to demonstrate the use of CBPCs for both macroencapsulation and microencapsulation of hazardous wastes [2,7,17,18]. Table 2 summarizes key performance data from these studies.

Singh et al. (1998) demonstrated the encapsulation of four waste streams with CBPC including cyrofractured debris, lead bricks, lead-lined plastic gloves, and mercury-contaminated crushed light bulbs. The study was a bench-

Table 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)	Hg leachate untreated (mg/L)	Hg leachate treated (mg/L)
Singh et al. (1998)	MA	BP	Cyrofractured debris	1.2–3 gallons	35	5000–7000	1.81	NA	NA
Singh et al. (1998)	MA	BP	Lead bricks	NR	NR	5000–7000	1.8	NA	NA
Sing et al. (1998)	MA	BP	Lead-lined gloves	5 gallons	NR	5000–7000	1.8	NA	NA
Singh et al. (1998)	MI	BP	Hg-contaminated crushed light bulbs	5 gallons	40	5000–7000	1.8	0.200–0.202	<0.00004–0.00005
DOE (1999a)[a]	MI	BP	DOE surrogate wastes of nitrate salts and off-gas scrub solution	NR	58–70	1400–1900	1.7–2.0	540–650	<0.00004–<0.00005
Wagh et al. (2000)[a]	MI	BP	DOE ash (HgCl$_2$ at 0.5 wt.%)	100 g	NR	NR	NR	40	<0.00085
Wagh et al. (2000)[a]	MI	BP	Delphi DETOX (with 0.5 wt.% each HgCl$_2$, Ce$_2$O$_3$, Pb(NO$_3$)$_2$)	100 g	NR	NR	NR	138–189	<0.00002–0.01
Wagh et al. (2000)[a]	MI	BP	Soil (HgCl$_2$ at 0.5 wt.%)	100 g	NR	NR	NR	2.27	<0.00015
Wagh and Jeong (2001)[b]	MI	BP	DETOX Wastestream (HgCl$_2$ at 0.5 wt.%)	162–500 g	60–78	NR	NR	250[c]	0.0047–0.0151
Wagh and Jeong (2001)[b]	MI	BP	DETOX Wastestream (Hg at 0.5 wt.%)	162–500 g	60–78	NR	NR	250[c]	0.00719–0.00764

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

scale project with waste form sizes ranging from 1.2 to 5 gallons and consequently some material handling and size reduction (e.g., shredding) was necessary. 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 potassium sulfide (K$_2$S) in the mixture for the mercury-contaminated crushed light bulbs. The mercury-contaminated crushed light bulbs were pretreated by mixing with a potassium sulfide solution for approximately 1 h. The glass was then set into CBPC with a formulation of 40 wt.% ash, 40 wt.% binder (MgO and KH$_2$PO$_4$ powders mixed in 1:1 molar ratio) and 20 wt.% water. 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 × 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 2 weeks prior to performance testing. 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 wastes and no gaps were present at waste–binder interfaces. TCLP tests on the mercury-contaminated wastes showed 200–202 µg/L in the untreated wastes compared to <0.04–0.05 µg/L for the treated wastes [2].

A U.S. Department of Energy study was completed to test the effectiveness of CBPCs in the treatment of salt-containing, mercury-contaminated mixed wastes [17]. 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 are soluble, easily hydrated, and cause deterioration of stability of the mineral microstructure over time by substitution reactions. As a consequence of these properties, salts adversely impact conventional cement matrices by causing a decrease in compressive strength and an increase in metal leachability. 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$_2$CO$_3$. These surrogate wastes were spiked with hazardous constituents including lead, chromium, mercury, cadmium, nickel, and trichloroethylene (TCE) at levels up to 1000 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. The CBPC binder was amended with K$_2$S, which successfully stabilized mercury to meet the TCLP limit in these wastes. However, wastes containing relatively high concentrations of salts (>42 wt.% salt loading) may leach Resource Conservation and Recovery Act (RCRA) hazardous metals, and could therefore compromise the long-term stability of the encapsulated materials.

Wagh et al. (2000) discusses the results of bench-scale studies for the encapsulation of mercury-contaminated sur-

rogate wastes including DOE ash waste, secondary waste streams from the DETOX^{SM} wet oxidation process, and contaminated topsoil [7]. The surrogate waste streams were dosed with mercuric chloride ($HgCl_2$) at 0.1–0.5 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. Long-term (90-day) leaching tests indicated that the diffusion of mercury through the CBPC matrix is 10 orders of magnitude lower than in cement systems.

Wagh and Jeong (2001) continued work related to the encapsulation of DETOX^{SM} wastes [18]. The study investigated with the effect of hematite (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.% Hg^0 and 95 wt.% Fe_2O_3. Two samples of each surrogate waste were pretreated with sodium sulfide nonahydrate ($Na_2S \cdot 9H_2O$) for 2 h, 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 3 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 Hg^0 wastes ranged from 7.19 to 7.64 µg/L. Waste loadings of 60–78 wt.% were achieved. Setting times were rapid (10–18 min) 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.%).

The following is a list of some of the reported advantages associated with the use of CBPC: (i) high concentration mercury wastes can be treated; (ii) lower temperature process (~176 °F) than SPSS and polyethylene encapsulation; (iii) no water removal is necessary as CBPC can be used to treat dry solids and sludges/liquids; (iv) unlike SPC, CBPC re-
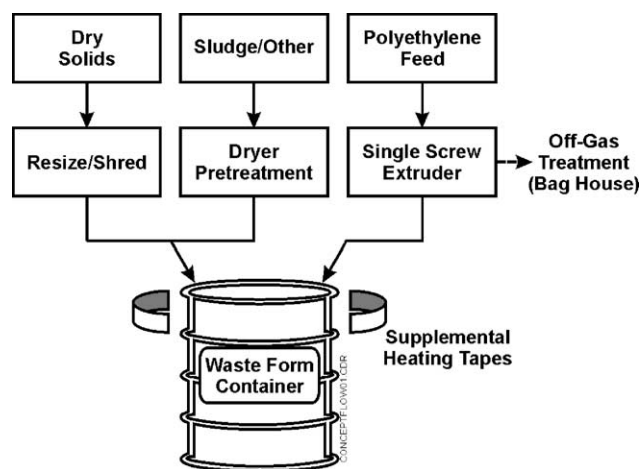

Fig. 3. Polyethylene macroencapsulation.

quires no additional heat input; (v) superior water tightness and chemical resistance compared to Portland cement; (vi) simple to implement since mixing and pouring equipment is readily available; (vii) nonflammable materials and stable and safe with oxidizing salts; (viii) does not generate secondary wastes or potentially hazardous off-gasses [2,7,18].

Based on the information reviewed from the sources previously identified, the following are the limitations of the process: (i) pretreatment with K_2S or other compounds is needed for chemical stabilization of mercury; CBPC alone is not enough; (ii) excess sulfide will increase the leachability of mercury, so careful processing is needed; (iii) some waste constituents (e.g., hematite) may accelerate setting times and decrease workability of the CBPC slurry; (iv) only limited data is available to support the long-term effectives and durability of CBPC waste forms; (v) the leaching of salt anions over time could deteriorate the integrity of the waste for high salt wastes.

The relative merits of the CBPC process compared to the SPSS process are discussed in the future development and research needs section.

8. Polyethylene encapsulation

Polyethylene is a thermoplastic material or a noncross-linked linear polymer that melts and liquefies at a specific transition temperature (248 °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 and Colombo (1997) the physical properties of LDPE are better suited to encapsulation because HDPE requires greater temperatures and pressures during processing and mixing with wastes [19].

Fig. 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. Kinetic mixers have also been used for polyethylene encapsulation [20]. Polyethylene macroencapsulation typically involves the use of a basket placed inside a drum to allow at least a 1 in. barrier around the waste material. Molten polyethylene is then poured from an extruder over and around the waste in the drum. In addition, an alternative to on-site pouring is the use of premanufactured containers as discussed below. Polyethylene microencapsulation typically involves directly mixing the waste material and polyethylene at an elevated temperature (typically 248–302 °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. 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 [21]. Both 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 tonnes 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 1–2 in. [20].

Several studies have been carried out using polyethylene for both macroencapsulation and microencapsulation of hazardous wastes [21–24]. In addition, several commercial vendors (e.g., Chemical Waste Management, Boh Environmental, and Ultra-Tech, International) provide macroencapsulation services using premanufactured HDPE containers. Only one study was found which dealt with the polyethylene microencapsulation of two types of mercury-contaminated wastes [23]. In general, there is little to no performance data available on the effectiveness of polyethylene encapsulation for mercury-contaminated wastes. Table 3 summarizes key performance data from several polyethylene encapsulation case studies (both with and without mercury wastes).

Burbank and Weingardt explored the use of polyethylene for the microencapsulation of mixed wastestreams at the DOE site in Hanford, Washington. Two wastes were tested that had relatively low concentrations of mercury, along with other heavy metals. An ammonium sulfate cake waste contained 9.2 ppm of mercury and a solar evaporation basin sludge contained 1.3 ppm of mercury. These wastes were incorporated into polyethylene at a 40–50 wt.% loading. Prior to encapsulation, calcium oxide was added to the higher waste loading (50 wt.%) specimens to help reduce metal leachability. However, the calcium oxide amendment did not reduce, but actually increased mercury leachability. The ammonium sulfate cake waste microencapsulated with polyethylene alone had a mercury TCLP of 0.442 mg/L. With the addition of calcium

oxide, this same waste stream had a higher mercury TCLP of 1.07 mg/L. It is clear from these results that polyethylene encapsulation alone cannot adequately reduce the availability or leachability of mercury, even at relatively low concentrations [23].

Also, due to the high processing temperatures of polyethylene encapsulation, it is likely that a large fraction of mercury would be volatilized unless it was pretreated or chemically fixed. This issue is highlighted by work completed by Carter et al. (1995) with arsenic, which is also a highly volatile metal. Carter et al. (1995) used HDPE with a melting point of 266 °F and an operating range at (356–410 °F) to microencapsulate powdered arsenic trioxide (As_2O_3). It was found that at a 20 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 with calcium carbonate, the volatility decreased, but achievable waste loadings in HDPE remained low. Mercury and its compounds are also highly volatile (e.g., mercuric chloride sublimes at 572 °F), so the results of this study provide some insight into the challenge of using polyethylene to process wastes containing high levels of either arsenic or mercury [24].

In addition to on-site processing, there are several vendors that provide macroencapsulation services with premanufactured HDPE containers including Chemical Waste Management, Boh Environmental, and Ultra-Tech, International. In general, the use of a tank or container is not considered macroencapsulation. However, premanufactured containers can be used and are allowed for contaminated debris under the LDR alternative debris standards at 40 CFR 268.45. Premanufactured containers can result in a reduction of the overall waste form volume by as much as one-fourth compared to an on-site pour of polyethylene [25].

Chemical Waste Management provides HDPE-lined roll-off boxes and 1/2-in.-thick HDPE vaults measuring 21 ft. × 7 ft. The vaults have a lid that is secured with adhesives and screws. Boh Environmental's Arrow-Pak[TM] technology consists of compacting 55-gallon drums filled with mixed or hazardous waste debris into 12-in.-thick pucks. The compacted drums are loaded into an 85-gallon metal overpack drum and then into a 1-in.-thick HDPE tube. Ultra-Tech, International offers a series of premanufactured, medium-density polyethylene containers. The containers can be custom-made in any size. 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 [26].

The following is a list of some of the advantages associated with the use of polyethylene as reported by previously identified researchers: (i) polyethylene has a high mechanical strength, flexibility, and chemical resistance; (ii) polyethy-

Table 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)	Hg leachate untreated (mg/L)	Hg leachate treated (mg/L)
Faucette et al. (1994)	MA	LDPE	BP/F	Combustibles, laboratory glassware, scrap metals, and lead (e.g., sheet, bricks, tape)	5–10 gallons	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 gallons	40–50[a]	1088–2465	NR	0.46 [9.2 ppm][b]	0.442–1.07
Burbank and Weingardt (1996)	MI	LDPE	BP	Solar basin sludge	1.25 gallons	40–50	1088–2465	NR	0.065 [1.3 ppm][b]	0.107–0.122
Carter et al. (1995)	MI	HDPE	BP	As$_2$O$_3$ with and without CaCO$_3$	NR	20 vol.%	NR	NR	NA	NA
Kalb et al. (1996)	MI	LDPE	BP	Off-gas scrub solution	NR	50–70	1950–2180	1.21–1.45	0.14	<0.009

[a] Prior to encapsulation, calcium oxide was added.
[b] Untreated waste TCLP not reported, estimated by total Hg level in waste divided by 20.

lene is readily available in post-consumer recycled forms; (iii) encapsulation equipment is commercially available and the process can be automated; (iv) premanufactured vaults and containers can be used under some circumstances; (v) polyethylene is used in landfill liners and extensive studies document its chemical resistance and long-term durability.

The following are some of the limitations of the process: (i) external heating is required and the process occurs at a higher temperature than the SPC and CBPC methods; (ii) the high temperatures make effective polyethylene encapsulation of mercury and arsenic problematic; (iii) polyethylene does not chemically incorporate the waste and mercury volatilization and leachability are a significant concern; (iv) wastes will typically have to be preprocessed to remove moisture and/or to achieve adequate particle size distributions.

The relative merits of the polyethylene encapsulation process compared to the SPSS and CBPC processes are discussed in the future development and research needs section.

9. Other encapsulation materials

Several other materials have been developed and demonstrated for the encapsulation of mercury-contaminated hazardous wastes including asphalt, polyester and epoxy resins, synthetic elastomers, polysiloxane, sol–gels (e.g., polyc-

erams), Dolocrete^{TM}, and carbon/cement mixtures. Key performance data from these studies are summarized in Table 4.

9.1. Asphalt

Asphalt or bitumen has been used to microencapsulate soil contaminated with low-levels of heavy metals [27,28]. Radian Corporation reported using cold-mix asphalt to microencapsulate soil contaminated with mercury (at 78 mg/kg). Hot-mix asphalt was deemed to be inappropriate because the elevated temperatures could promote the volatilization of mercury [29]. 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 [14].

9.2. 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 1000 ppm level. With polyester resins, waste loadings of 50 wt.% were achieved

Table 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)	Hg leachate untreated (mg/L)	Hg leachate treated (mg/L)
Kalb et al. (1996)	MI	Asphalt	BP	Off-gas scrub solution	NR	30–60	540–610	1.08–1.42	0.14	<0.009
Radian[a]	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	5100–6200	NR	50[b]	<0.01–0.2
Orebaugh (1993)	MA	Epoxy	BP	Mixed waste, lead billets	5 gallons	NR	NR	1.43–1.5 (resin only)	NA	NA
Carter et al. (1995)	MI	Styrene-butadiene rubber	BP	As$_2$O$_3$	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	0.034
DOE (1999b)	MI	Polyester resins	BP	Salt-containing mixed wastes	NR	50	5100–6120	1.03–1.09 (polyester resin only)	NR	<0.01–0.2
DOE (1999c)	MI	Poly-siloxane	BP	Salt-containing mixed wastes	NR	50	420–637	NR	50[b]	0.01–0.06
DOE (1999d)	MI	Sol–gels	BP	Salt-containing mixed wastes	NR	30–70	150–1500	NR	50[b]	0.044–0.23
Dolomatrix (2001)	MI	Dolocrete^{TM}	F	Hg-waste at 15,300 mg/kg	NR	NR	145	NR	765[b]	<0.1
Zhang and Bishop (2002)	MI	Powder reactivated carbon and cement	BP	Hg-contaminated sand up to 1000 mg/kg	NR	50	NR	NR	~0.10 to 10	~0.010 to 0.090

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

for unconcentrated spent off-gas scrub solutions and 70 wt.% for nitrate/chloride salts. Mixed waste, salt surrogate TCLP tests for mercury ranged from <0.01 to 0.2 mg/L [30]. In addition, Orebaugh (1993) has reported using several epoxy resins (e.g., Stycast 2651 and Thermoset 300) to macroencapsulate mixed waste, lead billets. However, no data on mercury encapsulation with epoxy resins was noted [31].

9.3. 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. Meng et al. (1998) reports using tire rubber for the treatment of mercury-contaminated soils [32]. 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 2–4% sulfur and less than 32% carbon black. Other researchers have reported using styrene-butadiene rubber (Solprene 1204) for the en-

capsulation of powdered arsenic trioxide (As$_2$O$_3$). Up to 64 wt.% of arsenic trioxide was incorporated into the rubber, but beyond this level the rubber became unworkable [24].

9.4. 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 °C or 86 °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 1000 ppm levels. Up to 50 wt.% waste loading was demonstrated. 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 [33].

9.5. Sol–gels

Sol–gels or polycerams are a hybrid material derived from the chemical combination of organic polymers and inorganic ceramics. A DOE study 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 1000 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 151 to 158 °F. Waste loadings from 30 to 70 wt.% were demonstrated. 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 [34].

9.6. Dolocrete^{TM}

Dolocrete^{TM} is a proprietary calcined dolomitic binder material that can be used for the microencapsulation of inorganic, organic, and low-level radioactive waste. Dolocrete^{TM} is reported to successfully encapsulate wastes containing aluminum, antimony, arsenic, bismuth, cadmium, chromium, copper, iron, lead, mercury, nickel, tin, and zinc. Mercury-contaminated wastes with up to 15,300 mg/kg have been reported to reach a final TCLP level of <0.1 mg/L [35].

9.7. Carbon and cement mixtures

Zhang and Bishop (2002) report using powdered reactivated carbon (PAC), along with Portland cement, to successfully encapsulate mercury-contaminated wastes. Surrogate wastes were created with up to 1000 mg/kg of mercury using sand, water, and $Hg(NO_3)_2$. These wastes were mixed with PAC and then solidified with Portland cement. The wastes were successfully treated to below the U.S. EPA TCLP limit for mercury. In addition, it was determined that pretreating the PAC with CS_2 increased its adsorption capacity for mercury by a factor of 10–100 times depending upon pH conditions. The authors report that this approach is a potentially cleaner and more effective means of stabilizing mercury wastes compared to sulfide precipitation [36].

10. 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-contaminated 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-contaminated hazardous wastes as discussed below.

Although several studies were noted which demonstrated the successful encapsulation of high-level, mercury-contaminated 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. It appears that SPC and CBPC are the only commercially viable encapsulation technologies for high concentration and/or elemental mercury wastes. 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. Polyethylene encapsulation is likely limited to the handling of debris and other wastes containing only minimal or low-levels of mercury. The Envirocare facility in Utah does have a full-scale system in place for polyethylene encapsulation. In addition, the use of premanufactured HDPE containers for macroencapsulation, as allowed under the U.S. EPA alternative debris standards, appears to offer a cost-effective disposal solution.

There are several technical issues related to the implementation of SPC and CBPC encapsulation that need to be resolved. A better understanding of the long-term stability of the final waste forms may be needed. In general, the long-term stability of materials encapsulated with SPC or CBPC has not been addressed and waste form degradation may be promoted by high salt loadings in wastes and other factors. An improved understanding is needed of the kinetics of low-temperature processes such as SPC or CBPC. This additional information could help in scale-up and process optimization. CBPC is prone to rapid setting in the presence of reactive wastes (e.g. hematite) and rapid cooling with SPC can lead to the formation of undesirable air pockets. For all technologies that rely upon fixing mercury into its mercuric sulfide form, there is a need to further assess 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.

The use of other innovative materials (e.g. synthetic elastomers, polyester resin, Dolocrete^{TM}, etc.) 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.

It is clear that waste specific treatability tests will be required for the selection of the most appropriate encapsulation

material for a given industrial waste stream. 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.

Acknowledgements

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