

# Sulfurization and Solidification of Wastes Consisting of Elemental Mercury

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## Abstract

Entry into force of the Minamata Convention on Mercury has changed the supply-demand balance, leading to an oversupply of mercury worldwide. We assessed sulfurization and solidification technologies for wastes consisting of elemental mercury for disposal in specially engineered landfills. For chemical stabilization of elemental mercury, gas phase sulfurization, wet sulfurization in water phase and dry mechanochemical sulfurization were evaluated. Solidification for sulfurized mercury was examined using cement/mortar/concrete, dicyclopentadiene-modified sulfur and epoxy resin as solidification materials. In this research, crucial environmental requirements (limits of leaching and emission) for sulfurized and solidified products were defined and evaluated using three Japanese leaching tests (JLT-13, JLT-46 and a tank leaching test) and a headspace test. Overall, each sulfurization and solidification technology fulfilled the crucial environmental requirements for sulfurized and solidified products and compressive strength of the solidified products.

**Key words :** mercury wastes, Minamata Convention on Mercury, multiple protection, solidification, sulfurization

## 1. Introduction

The Minamata Convention on Mercury entered into force on 16 August, 2017 to protect human health and the environment from anthropogenic emissions and releases of mercury and mercury compounds (Art. 1) (UNEP, 2019). Mercury wastes are substances or objects in a quantity above the relevant thresholds and should be managed in an environmentally sound manner (ESM), taking into account the Basel Technical Guideline and requirements in an additional annex adopted by the Conference of Parties (COP) (Art. 11, para. 2 & 3) (UNEP, 2019). The entry into force of the Convention has changed the supply-demand balance worldwide, notably decreasing the demand for mercury. Inevitably, there will be an excess of mercury recovered from mercury wastes and industrial processes. Herein, the amounts of excess mercury worldwide and in Japan are estimated to reach 24,000 to 25,000 and 600 to 1,400 metric tons, respectively, by 2050 (Sodeno & Takaoka, 2017). A variety of concepts, considerations, operations and combinations of these are available depending on the type and source of mercury wastes and the situation of each country. As an example, long-term storage, not disposal, of commodity grade mercury in warehouses with safety measures is ongoing at the Hawthorne Army Depot in

Nevada, U.S. (UNEP, 2015). On the other hand, mercury waste is being disposed of for permanent storage in underground salt mines at depths of 500 to 800 meters away from the biosphere, *e.g.*, Herfa-Neurode in Hessen, with natural and artificial barriers in Germany (UNEP, 2015). Japan has also taken a defense-in-depth approach, which is purification, stabilization and solidification of mercury wastes similar to Spain's sulfur polymer stabilization/solidification (SPSS) (Fuhrman *et al.*, 2002). The stabilized and solidified mercury is disposed of in a specially engineered landfill (SEL), which has been mandated by an enforcement order of the Waste Management and Public Cleansing Law since October, 2017 in Japan.

The recovery (purification) of elemental mercury from mercury wastes is performed through thermal treatment, *e.g.*, a multiple-hearth furnace (Murase & Matsubara, 2013) or rotary kiln (Hawk & Aulbaugh, 1998), due to its low boiling point, 356.73°C/629.88 K (Lide, 1999). The elemental mercury kicked out from the wastes into the gas phase is recovered in a cooled condenser and further purified by successive distillation.

Chemical stabilization of recovered elemental mercury is plausible because of its unique nature, the only metal that exists as a liquid at room temperature and low boiling point. Elemental mercury is chemically

stabilized via sulfurization, selenization, amalgamation, etc. (Hagemann, 2009). Sulfurization of metallic mercury with sulfur powder proceeds spontaneously at room temperature. Mercury sulfide (cinnabar,  $\alpha$ -HgS and metacinnabar,  $\beta$ -HgS) is a stable and water-insoluble mineral, and an increase of excess sulfur worldwide is also expected due to IMO regulation on SO<sub>x</sub> emissions from fuel oil (IMO, 2016). Mercury selenide (tiemannite, HgSe) is also a very stable and water-insoluble mineral, but its formation reaction cannot proceed at room temperature and the toxicity of selenium itself is a concern. Mercury can make an alloy (amalgam) with several metals, *e.g.*, zinc, copper or gold. However, the vapor pressure of mercury of those amalgams is not so low even after amalgamation compared to that of elemental mercury, and some of the counter metals are expensive. Accordingly, sulfurization has been the main technology adopted for chemical stabilization. We investigated the feasibility of gas phase sulfurization of elemental mercury and also wet sulfurization in water for a mercury compound compared to our past research on dry mechanochemical sulfurization using a planetary ball mill for elemental mercury (Fukuda *et al.*, 2014 and Nakamura *et al.*, 2017).

Cement/mortar/concrete (Nakayama *et al.*, 2015), asphalt/bitumen (Wati *et al.*, 1992) and thermoplastic/thermosetting resins (Ito *et al.*, 1984) have been widely used as solidification materials for radioactive waste with pros and cons, and they are also thought to be applicable to sulfurized mercury. Modified sulfur, synthesized by modifying sulfur with dicyclopentadiene (DCPD) etc., was adopted in Japan as the sole solidification material in the Guidelines on Mercury Wastes to satisfy certain testing standards, *e.g.*, compressive strength and leachability, (Ministry of the Environment, 2017). We have been developing a solidification method using a thermosetting and impervious epoxy resin for sulfurized mercury comparable to DCPD-modified sulfur solidification (Uenishi *et al.*, 2018). The relevance of

solidification of sulfurized mercury using cement, DCPD-modified sulfur and epoxy resin as solidification materials was investigated at the laboratory scale. The objective herein was to demonstrate our laboratory-scale sulfurization and solidification technologies for wastes consisting of elemental mercury. A cement/mortar/concrete solidification method was newly developed in this research, and other methods for sulfurization and solidification were developed and are summarized here.

## 2. Materials and Methods

### 2.1 Sulfurization

#### 2.1.1 Gas Phase Sulfurization

The reaction between mercury (99.5%, Wako) and sulfur (98%, Nacalai Tesque) in the gas phase proceeded in a vertical tubular electric furnace (FT-POT300-VAC, Full-Tech) at high temperatures (Fig. 1). The inside dimensions of the furnace were 20 cm D×40 cm H (*ca.* 12.6 L). This gas phase sulfurization was examined by optimizing operating factors such as input molar ratio (S/Hg) of sulfur to mercury (1.05, 1.25, 1.50), temperature (500 °C and 600 °C) and reaction time (0.5 h and 1.0 h) (Hamaguchi *et al.*, 2013). The initial input mercury amount was set at 40.00 g, and the internal total and O<sub>2</sub> partial pressures were  $1.5 \times 10^3$  Pa and 100 ppm O<sub>2</sub>, respectively. After the reaction, the gas inside the vessel was drawn from the gas outlet of the vessel into an impinger filled with 1 M NaOH (as an SO<sub>2</sub> absorber), following two impingers with 0.01 M KMnO<sub>4</sub> with 3% H<sub>2</sub>SO<sub>4</sub> using a suction pump at 0.9 L/min for 22 min. After trapping, the mercury concentration in the KMnO<sub>4</sub> solutions was determined according to JIS K 0222.

#### 2.1.2 Wet Sulfurization in the Water Phase

The sulfurization of mercuric chloride (99.5%, Wako) proceeded with sodium sulfide (98%, Wako) in a 5-L stainless beaker. HgCl<sub>2</sub> was dissolved into 4.00 L of pure water or 0.1 mol/L sulfuric acid solution with an initial mercury concentration of 1,000 mg Hg/L.

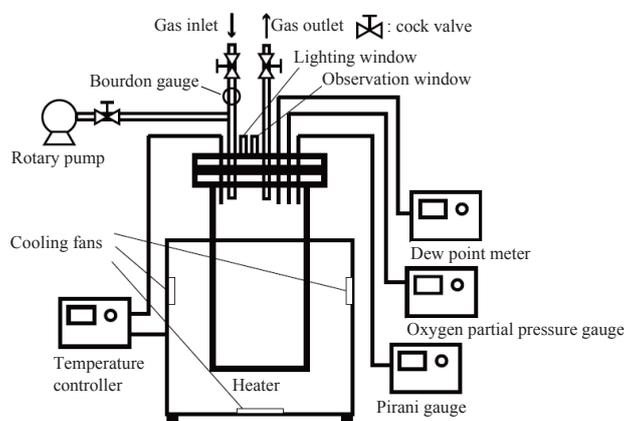


Fig. 1 Vertical tubular electric furnace for gas phase sulfurization.

Following pH adjustment to 7.0 using NaOH or Ca(OH)<sub>2</sub>, a 10w/v% Na<sub>2</sub>S solution was added at a molar ratio (S/Hg) of 1.10, and then the excess sulfide was removed by adding a 10w/v% solution of iron (II) sulfate (99%–102%, Wako). Coagulation was performed to collect the sulfurized products in the water phase using a 10w/v% aluminum sulfate (>85%, Wako) as a coagulant. Durations of rapid- and slow-mixings and settlement were 3, 15 and 10 min respectively. The final products were obtained by 1.0- $\mu$ m membrane filtration and dried at 60°C for 48 h (Miyahara *et al.*, 2017).

### 2.1.3 Dry Mechanochemical Sulfurization

A mechanochemical reaction between elemental mercury (99.99%, Nomura Kohsan) and sulfur powder (98%, Nacalai Tesque) was attained using a planetary ball mill (BX382, Kurimoto) with two reaction vessels and 25-mm balls. The sulfur powder was added to 1,152 g of elemental mercury at a molar ratio (S/Hg) of 1.05 in a 2,400-mL vessel, accordingly 2,304 g of metallic mercury per batch. The operating conditions were ball filling ratio: 30%, centrifugal acceleration: 30×*G* and reaction time: 60 min (Nakamura *et al.*, 2017). Dry mechanochemical sulfurization was conducted eleven times (i.e., in 22 vessels) to evaluate its repeatability.

Sulfurized mercury for examination of cement/mortar/concrete solidification was synthesized using a planetary mono mill (Pulverisette-6, Fritsch). The leaching value by Japanese leaching test 46 (JLT-46) and mercury concentration in the headspace were 0.06  $\mu$ g/L and 1.0  $\mu$ g/m<sup>3</sup>, respectively (Fukuda *et al.*, 2014).

## 2.2 Solidification

### 2.2.1 Cement, Mortar and Concrete Solidifications

The sulfurized mercury was solidified using a cement, mortar and concrete. Ordinary Portland cement (Taiheiyo Cement), sand as a fine aggregate (Japan Cement Association) and gravel as a coarse aggregate (Yura River, Japan) were used as solidification materials and mixed with sulfurized mercury and water using a

mixer (KC-8, Kansai Kiki) at a water-cement ratio (w/c) by weight of 50% (Table 1). Fresh mix was poured into a mold (50 mm D×100 mm H) and immediately placed in a temperature and humidity-controlled chamber (25°C and 60%RH) for 24 h. After demolding, membrane curing was conducted for the cylindrical specimens for 55 days. Materials recovered after a compressive strength test were put through a Japanese standard leaching test, JLT-13, within a few days.

### 2.2.2 Modified Sulfur Solidification

Sulfur powder was melted at 135 °C for 10 min in a mixer (HIVIS MIX 2P-1, PRIMIX), and modified by adding dicyclopentadiene (DCPD) as a modifier to 5wt%. After replacement of air by nitrogen gas inside a vessel, the modification reaction proceeded by mixing at 50 rpm and 135°C for 1 h. The modified sulfur was cooled at room temperature for 24 h and then powdered using a stamp mill (ANS-143, Nitto Kagaku).

The modified sulfur was melted in the mixer at 130°C for 10 min and mixed with sulfurized mercury at a weight ratio of sulfurized mercury to modified sulfur of 50%. After gas replacement as before, the ingredients were mixed at 50 rpm and 130°C for 1 h. The mixture was poured into the mold and allowed to cool at room temperature for 24 h. (Uenishi *et al.*, 2018). When transferring the melted product from the mixer to the mold, the mercury concentration in the working environment was directly monitored using a portable mercury survey meter (EMP-2, Nippon Instruments).

### 2.2.3 Epoxy Resin Solidification

Two bisphenol A (BPA) epoxy resins and four curing agents (amines) were selected to explore the optimum mixing ratio, eight combinations in total, in this research. A liquid epoxy resin (jER 828) and a curing agent (jER ST12) were purchased from Mitsubishi Chemical. A low-viscosity liquid epoxy resin (diluted jER 828) and curing agents (J-882, B-2413, X-7000) were obtained from Daito Sangyo. The mix ratios were calculated based on the epoxide equivalent weight (EEW) of the epoxy resins

**Table 1** Mix proportions for cement, mortar and concrete solidifications.

Material	w/c	HgS (%)	HgS (g)	Cement (g)	FA (g)	CA (g)	Water (g)
Cement	0.50	20	200	800	-	-	400
		40	400	600	-	-	300
		60	300	200	-	-	100
	0.60	20	200	800	-	-	480
		40	400	600	-	-	360
		60	600	400	-	-	240
Mortar	0.50	5	60	400	740	-	200
		10	120	400	680	-	200
		20	240	400	560	-	200
Concrete	0.50	5	75	250	425	750	125
		10	150	250	350	750	125

w/c: a water-cement ratio by weight, FA: fine aggregate, CA: coarse aggregate.

and the amine hydrogen equivalent weight (AHEW) of the curing agents. The epoxy resin, sulfurized mercury and curing agent were mixed in a plastic beaker by hand or agitator (BLh300, Shinto Scientific) at weight ratios of sulfurized mercury of 0%, 50%, 60%, 70%, 80% and 90% against the epoxy resin and curing agent. The 48 combinations (eight epoxy resins and six weight ratios of sulfurized mercury) were finally examined. The mixtures were poured into the mold and cured at 40°C for 16 h (Uenishi *et al.*, 2018).

## 2.3 Testing and Analytical Methods

### 2.3.1 Compressive Strength

The testing method for the compressive strength of concrete (JIS A 1108:2018) was applied to specimens solidified in the mold (50 mm D×100 mm H). A minimum compressive strength of 0.98 MPa is required as a landfill standard in Japan.

### 2.3.2 Japanese Standard Leaching Tests (JLT-13 and JLT-46)

The JLT-13 test is designed to evaluate the leachability from industrial wastes compared to landfill standards by a single batch extraction. Solidified specimens were pulverized into 0.5–5 mm and mixed with water at a liquid to solid ratio (L/S) of 10. Following shaking at 20 °C for 6 h, the liquid extracts were obtained by 1.0- $\mu\text{m}$  membrane filtration and the dissolved mercury concentration determined using a reducing-vaporization mercury analyzer (RA-4300, Nippon Instruments). The landfill standard for mercury and its compounds is 5  $\mu\text{g/L}$  or less in Japan.

JLT-46 is a test method to judge whether the environmental standard for soil is met or not. JLT-46 is similar to JLT-13, but the sample size (less than 2 mm) and pore size (0.45  $\mu\text{m}$ ) of the membrane filter differ. Japanese environmental standard for mercury and its compounds in soil is 0.5  $\mu\text{g/L}$  or less.

### 2.3.3 Headspace Test

The mercury concentration in the headspace above the samples (sulfurized mercury and sulfurized/solidified mercury) was determined using continuous mercury analyzers (MS-1A/DM-6B or WLE-8/EMP-2, Nippon Instruments) (Fukuda *et al.*, 2014). The headspace concentration of mercury for stabilized/solidified mercury wastes is not regulated in Japan, so the provisional regulatory standard of the EU (3  $\mu\text{g/m}^3$ )—the limit of detection of a used analytical instrument (BiPRO, 2010)—and the working environment evaluation standard for mercury in Japan (25  $\mu\text{g/m}^3$ ) were referred to as temporal criteria in this research.

### 2.3.4 Tank Leaching Test

The Japanese tank leaching test is a diffusion test to evaluate the leaching behavior from monolithic specimens using a single static-tank leaching procedure (Ministry of Construction, 2000). After allowing the tank to stand at 20 °C for 28 days, the mercury concentration

in the 0.45- $\mu\text{m}$  membrane filtered samples was determined using the RA-4300.

### 2.3.5 Powder X-ray Diffraction (XRD) and Rietveld Refinement

The XRD (Cu- $K_{\alpha}$ ) pattern of the sulfurized products was obtained using an X-ray diffractometer (RINT-UltimaPC, Rigaku), and its Rietveld refinement was conducted to quantitatively calculate the percentage of cinnabar and metacinnabar in the sulfurized mercury.

## 3. Sulfurization

### 3.1 Gas Phase Sulfurization

Recovered mercury ranged from 60%–70% on average, and sulfurized mercury existed mainly as cinnabar in the products of gas phase sulfurization. The results of gas phase sulfurization are shown in Tables 2 and 3 (Hamaguchi *et al.*, 2013). When the mix ratio of S/Hg was set to 1.05 and 1.50 at 500/600 °C for 1.0 h, the resulting products satisfied the environmental standard for soil under JLT-46. However, the case in the condition of S/Hg = 1.25 did not satisfy it. The mercury concentrations in the headspace of the resulting products were below the EU's provisional regulatory standard (3  $\mu\text{g/m}^3$ ) when S/Hg was 1.25 at 500/600°C and when S/Hg was 1.50 at 500°C for 1.0 h, whereas the others exceeded the standard. As a result, our gas phase sulfurization was a little unstable, but the properties of the resulting products were comparable to DELA's assessment, in which the leaching value was less than 50  $\mu\text{g/L}$  (9  $\mu\text{g/L}$  on average) by German DEV S4/ DIN 38 414-4 and the mercury vapor pressure was below 3  $\mu\text{g/m}^3$  (Hagemann, 2009).

The mercury concentration within the vessel after the reaction (157–389  $\mu\text{g/m}^3$ ) was one order of magnitude

**Table 2** Leaching values ( $\mu\text{g/L}$ ) of resulting products in gas phase sulfurization by JLT-46 (mean  $\pm$  SD,  $N = 3$ ).

	Reaction time (h)	0.5		1.0	
		500	600	500	600
S/Hg	1.05	8.42 $\pm$ 0.63	8.75 $\pm$ 0.20	<u>0.32 <math>\pm</math> 0.00</u>	<u>0.24 <math>\pm</math> 0.01</u>
	1.25	7.86 $\pm$ 0.04	6.62 $\pm$ 0.14	1.23 $\pm$ 0.06	0.68 $\pm$ 0.05
	1.50	6.10 $\pm$ 0.34	4.25 $\pm$ 0.02	<u>0.06 <math>\pm</math> 0.00</u>	<u>0.20 <math>\pm</math> 0.00</u>

Underlines indicate values below the environmental standard (0.5  $\mu\text{g/L}$ ).

**Table 3** Mercury emissions ( $\mu\text{g/m}^3$ ) from resulting products in gas phase sulfurization.

	Reaction time (h)	0.5		1.0	
		500	600	500	600
S/Hg	1.05	32.9	39.9	35.8	56.9
	1.25	54.9	30.7	<u>1.6</u>	<u>0.8</u>
	1.50	4.3	9.8	<u>1.6</u>	21.0

Underlines indicate values lower than 3  $\mu\text{g/m}^3$ .

higher compared to the working environment evaluation standard for mercury in Japan ( $25 \mu\text{g}/\text{m}^3$ ). Under these circumstances, leakage of mercury from the vessel becomes a big concern in gas phase sulfurization.

### 3.2 Wet Sulfurization

Mercuric ion ( $\text{Hg}^{2+}$ ) with an initial concentration of 1,000 mg as Hg/L was quantitatively recovered as a mercury sulfide through a wet sulfurization process in the conditions of both pure water and dilute sulfuric acid (0.1 mol/L). XRD analysis demonstrated that the resulting products were a metacinnabar showing low crystallinity compared to that by dry mechanochemical sulfurization (Fukuda *et al.*, 2014). The leaching values of all resulting products from wet sulfurization satisfied the JLT-13 standard ( $5.0 \mu\text{g}/\text{L}$ ) and those mercury emissions were below  $0.1 \mu\text{g}/\text{m}^3$  (Miyahara *et al.*, 2017). A drying process is additionally necessary due to high water content (*ca.* 80%) of the products. In the case of dilute sulfuric acid as a solvent, the gypsum content as a by-product exceeded 90% in the products, which is undesirable from the viewpoint of volume reduction.

### 3.3 Dry Mechanochemical Sulfurization

All resulting products in dry mechanochemical sulfurization satisfied the JLT-46 standard ( $0.5 \mu\text{g}/\text{L}$  or less), and the mercury concentration in the headspace was  $1.7 \pm 1.5 \mu\text{g}/\text{m}^3$  (mean  $\pm$  SD), with 18 out of 22 samples (86%) satisfying the EU's provisional regulatory standard ( $3 \mu\text{g}/\text{m}^3$ ). XRD-Rietveld analysis demonstrated the percentage of cinnabar, which is a relatively more stable crystal than metacinnabar, in the sulfurized mercury was 48.6%.

Overall, dry mechanochemical sulfurization can stabilize elemental mercury with high repeatability. Calculated under conditions of five days/week and 52 weeks/year, three operations per day (6.9 kg/day at optimum conditions) could give an annual treatment capacity of *ca.* 1,800 kg/year even with the small pilot mill machine.

As a conclusion so far, dry mechanochemical sulfurization has advantages and greater reliability than the gas phase and wet sulfurization processes.

## 4. Solidification

Mercury sulfurized by dry mechanochemical sulfurization was used for examining solidification methods. The results of JLT-13 and the headspace test for sulfurized mercury were below  $5.0 \mu\text{g}/\text{L}$  and  $3 \mu\text{g}/\text{m}^3$ , respectively.

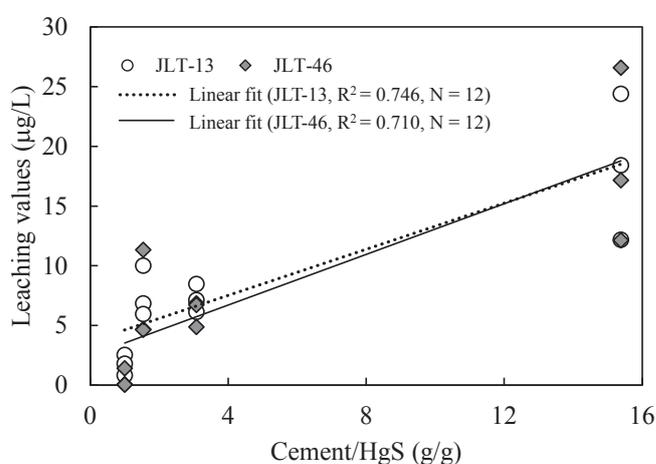
### 4.1 Cement, Mortar and Concrete Solidification

The results of compressive strength, JLT-13 and tank leaching test for cement/mortar/concrete solidified products are summarized in Table 4. One mortar-

**Table 4** Compressive strength and leaching values of cement, mortar and concrete solidified products.

Material	w/c	HgS (%)	Compressive strength (MPa)	Leaching value ( $\mu\text{g}/\text{L}$ )	
				JLT-13	Tank leaching
Cement	0.50	20	46.2	0.34	0.16
		40	27.1	0.20	0.25
		60	31.3	0.11	0.06
	0.60	20	18.3	0.25	0.10
		40	18.2	0.32	0.12
		60	16.3	0.27	0.22
Mortar	0.50	5	34.0	0.24	0.96
		10	37.7	0.34	0.26
Concrete	0.50	20	39.3	0.38	3.41
		5	20.8	0.10	0.16
		10	27.0	0.07	0.05

w/c: water-cement ratio by weight. Compressive strength was evaluated at a material age of 56 days.



**Fig. 2** Relationship between the leaching behavior of mercury under JLT-13/-46 and the mix ratio of high early strength Portland cement to sulfurized mercury from a preliminary examination. Specimens solidified with concrete (HgS = 1, 5, 10 wt%) and cement (HgS = 40 wt%) with a water/cement ratio of 0.50 were cured in water for a week.

solidified specimen (HgS, 20%) exhibited high leaching values in the tank leaching test, whereas all the solidified specimens including mortar-solidified products satisfied landfill standards ( $5 \mu\text{g}/\text{L}$  or less) under JLT-13. All of the cement- and concrete-solidified products satisfied the standards, but the sulfurized mercury content of the concrete-solidified specimens was relatively lower than that of the cement-solidified ones. Our preliminary examination of cement and concrete solidification using a high early strength Portland cement (Sumitomo Osaka Cement) demonstrated a positive correlation between the leaching behavior of mercury under JLT-13/-46 and the mix ratio of cement to sulfurized mercury (cement/HgS by weight) (Fig. 2). This is consistent with the fact that the solubility of HgS drastically increases under alkaline conditions ( $\text{pH} > 10$ ) (Kadotani *et al.*, 2009), which suggests that if the mixture of solidification material(s) and sulfurized mercury can be cured to achieve the

requirements, a lower ratio of cement would be preferable to suppress mercury leaching.

#### 4.2 Modified Sulfur Solidification

Solidification of sulfurized mercury with DCPD-modified sulfur at a weight ratio of 1:1 was conducted following the Guidelines on Mercury Wastes in this research. The results of compressive strength, JLT-13 and headspace test for sulfurized/solidified mercury with modified sulfur were 30.6 MPa, 0.05  $\mu\text{g/L}$  and  $< 0.1 \mu\text{g/m}^3$ , respectively, all of which satisfied standards (Uenishi *et al.*, 2018). The tank leaching test demonstrated lower leaching behavior ( $< 0.01 \mu\text{g/L}$ ) of mercury from a monolith solidified with DCPD-modified sulfur compared to that (0.05  $\mu\text{g/L}$ ) from pulverized monolith (0.5–5 mm) by JLT-13's protocol. Modified sulfur solidification is effective and successful at encapsulating sulfurized mercury. When opening the vessel, mercury vapor was detected at *ca.* 5.0  $\mu\text{g/m}^3$ , which is below the Japanese working environment evaluation standard (25  $\mu\text{g/m}^3$ ).

#### 4.3 Epoxy Resin Solidification

Following a screening examination based on JLT-13 and the headspace test, the optimum mix ratio was decided as follows: epoxy resin: jER 828, curing agent: B-2413, and weight ratio of sulfurized mercury: 80%. B-2413, 1,3-bis(aminomethyl)cyclohexane (1,3-BAC), is a highly-reactive cycloaliphatic diamine, which offers advantages in UV resistance and clarity owing to its lack of conjugated double bonds; heat resistance due to high glass transition temperature ( $T_g$ ), curing even at low temperature and high humidity; and cost saving through material reduction due to its low AHEW.

The results of compressive strength, JLT-13 and the headspace test for sulfurized mercury solidified with the epoxy resin were 107.0 MPa, 0.09  $\mu\text{g/L}$  and  $< 0.1 \mu\text{g/m}^3$ , respectively, all of which satisfied standards (Uenishi *et al.*, 2018). The tank leaching test showed lower leaching behavior ( $< 0.01 \mu\text{g/L}$ ) of mercury from a monolith solidified with the epoxy resin compared to that (0.09  $\mu\text{g/L}$ ) from a pulverized monolith (0.5–5 mm) under JLT-13's protocol. Epoxy resin solidification is also effective and successful at encapsulating sulfurized mercury.

The mercury concentration in the curing chamber increased up to 1.0  $\mu\text{g/m}^3$  at a maximum as the central temperature in the mold increased up to 150°C about 15 min after curing started, after which it decreased. When solidifying the same weight of sulfurized mercury, the volume of the monolith of epoxy resin was roughly half that of the DCPD-modified sulfur.

Seven epoxy resins other than the optimum one (jER 828 and B-2413) can still solidify sulfurized mercury at a weight ratio of 70% and satisfy standards. As a result, solidification of sulfurized mercury with epoxy resin has

advantages similar to or better than that with DCPD-modified sulfur.

## 5. Conclusions

A comprehensive examination was conducted to establish stabilization and solidification technologies for disposal of wastes consisting of elemental mercury in SELs. We conclude that dry mechanochemical sulfurization of elemental mercury using a planetary ball mill has advantages and reliability over gas phase sulfurization of elemental mercury and wet sulfurization of an inorganic mercury compound in the water phase. However, it is necessary to check whether the sulfurization reaction by the dry mechanochemical process is properly proceeding for each lot. The dry mechanochemical process has an annual treatment capacity of *ca.* 1,800 kg/year. Epoxy resin solidification has advantages similar to or better than DCPD-modified sulfur solidification, which has been adopted in the Guidelines on Mercury Wastes in Japan. We propose epoxy resin solidification as an alternative solidification technology. Further research will be required to evaluate and secure long-term stability of sulfurized and solidified mercury wastes in SELs.

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