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Treating high-mercury-containing lamps using full-scale thermal desorption technology

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ABSTRACT

The mercury content in high-mercury-containing lamps are always between 400 mg/kg and 200,000 mg/kg. This concentration is much higher than the 260 mg/kg lower boundary recommended for the thermal desorption process suggested by the US Resource Conservation and Recovery Act. According to a Taiwan EPA survey, about 4,833,000 cold cathode fluorescent lamps (CCFLs), 486,000 ultraviolet lamps and 25,000 super high pressure mercury lamps (SHPs) have been disposed of in the industrial waste treatment system, producing 80, 92 and 9 kg-mercury/year through domestic treatment, offshore treatment and air emissions, respectively. To deal with this problem we set up a full-scale thermal desorption process to treat and recover the mercury from SHPs, fluorescent tube tailpipes, fluorescent tubes containing mercury-fluorescent powder, and CCFLs containing mercury-fluorescent powder and monitor the use of different pre-heating temperatures and desorption times. The experimental results reveal that the average thermal desorption efficiency of SHPs and fluorescent tube tailpipe were both 99.95%, while the average thermal desorption efficiencies of fluorescent tubes containing mercury-fluorescent powder were between 97% and 99%. In addition, a thermal desorption efficiency of only 69.37-93.39% was obtained after treating the CCFLs containing mercury-fluorescent powder. These differences in thermal desorption efficiency might be due to the complexity of the mercury compounds contained in the lamps. In general, the thermal desorption efficiency of lamps containing mercury-complex compounds increased with higher temperatures.

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1. Introduction

Mercury, which has the lowest melting point $(-39 \circ C)$ of all the pure metals, is the only pure metal that is liquid at room temperature [1,2]. Mercury is also one of the most toxic chemicals and has been placed on the priority list of 129 hazardous chemical substances by the United States Environmental Protection Agency (USEPA). Mercury is a persistent environmental pollutant with bioaccumulation ability in fish, animals, and human beings. In the United States, there is already legislation aimed at controlling some mercury-containing products. The European Union proposed the control mercury-containing products in their "Council Directive 76/769/EEC". However, due to its several physical and chemical advantages such as its low boiling point ($357 \circ C$) and easy vaporization, mercury is still an essential material in many industrial products.

Chief among these mercury-containing industrial products are light sources, especially cold cathode fluorescent lamps (CCFLs), ultraviolet (UV) lamps, and high pressure mercury lamps (SHPs), which are produced and used worldwide. Mercury is used in fluorescent bulbs for converting electrical energy to radiant energy in the ultraviolet range and then re-radiating it in the visible spectrum [3]. However, the mercury concentration and the speciation in the above lamps can vary, depending on the manufacturer, lamp type and year of manufacturing [4]. According to a survey made by the USEPA, the mercury contained in one type of fluorescent lamp (produced by the Sylvania lamp manufacturer) probably consists of 0.2% (0.042 mg) elemental mercury (vapor phase) and 99.8% (20.958 mg) divalent mercury incorporated into the phosphorus powder. It is acknowledged that the speciation of mercury contained in fluorescent lamps is a controversial and complex subject [5]. Unfortunately, when such mercury-containing lamps are discarded, the mercury, which is harmful to human beings and other organisms, is released into the environment [6].

However, several treatment technologies, such as thermal/chemical treatment and solidification/stabilization, have been



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evaluated by the USEPA, depending on the mercury concentration in the mercury-containing waste. The Resource Conservation and Recovery Act (RCRA) of the United States suggests that mercurycontaining waste can be treated by solidification/stabilization when the mercury content is less than 260 mgHg/kg, while thermal desorption methods should be used when the mercury content is higher than 260 mgHg/kg [5].

In the past decade, Taiwan has produced huge amounts of high technology products. Such industrial products often include highmercury-containing lamps (e.g., CCFLs, UV and SHPs) as one of their components. According to a survey made by the Taiwan EPA, about 4,833,000 CCFLs lamps, 486,000 UV lamps and 25,000 SHPs have already entered the industrial waste treatment system. It was also calculated that 44%, 51% and 5% kg-mercury/year have been processed via domestic treatment, offshore treatment and emission in air, respectively [7]. The mercury content in the above lamp waste is from 400 mg/kg to 200,000 mg/kg, which is much higher than 260 mg/kg. Thus, a thermal adsorption process should be used to treatment and recovery of mercury-containing lamps, according to the suggestion of RCRA of United States.

Generally, the full-scale thermal desorption process needed to recover mercury from mercury-containing waste includes a pretreatment unit and a mercury recovery system. In the pretreatment unit thermal desorption technology is used to separate the mercury from the other constituents, after which the mercury vapor is further recovered by a condenser in the mercury recovery system. There are already several commercial thermal desorption technologies available internationally, including the MRT system of Sweden [8], the Nomura system of Japan, and the AERC, SepaDyne, and Mercury Recovery technologies of the United States [9-11]. Of these thermal desorption technologies, the Nomura system is a wet-based while the others are dry-based thermal desorption systems. There are several disadvantages to a wet-based thermal desorption system including the production of secondary hazardous pollutants (e.g., mercurycontaining sludge), the complex post-treatment process required for the wastewater, and high cost. In contrast, the advantages of the dry-based thermal desorption systems include low production of secondary waste, low cost and high economic efficiency.

In addition, there are no pretreatment facilities in the SepaDyne, and Mercury Recovery technology's thermal desorption systems to the crushing of the lamps. The AERC, Nomura, and MRT processes do have such pretreatment units. However, neither the AERC nor the Nomura systems are suited for use in Taiwan, because of their high maintenance cost and complex procedures, although they have mercury recovery efficiencies of 99.999% and 99.99%, respectively [2,8–11]. This is why, in this study, we use an MRT thermal desorption system to treat the mercury-contained lamps.

Desorption time and temperature are the key factors influencing the desorption efficiency of all thermal desorption processes [12]. Thus, in this study, a full-scale thermal desorption plant was set up to recover the mercury from high-mercury-containing lamps using different desorption times and temperatures.

2. Materials and methods

2.1. Full-scale thermal desorption process

In this study we treated SHPs, fluorescent tube tailpipes, mercury-fluorescent powder containing fluorescent tubes, and mercury-fluorescent powder containing CCFLs by a thermal desorption process to recover the mercury. Fig. 1 shows a schematic diagram of the full-scale thermal desorption process, which can be divided into a lamp crushing system and a thermal desorption system. The lamp crushing system includes the machines for crushing fluorescent powder containing lamps and airtight high pressure mercury lamp crushing machine, while the thermal desorption system includes the thermal desorption oven and mercury recovery system.

2.1.1. Lamp crushing system

Two lamp crushing machines were used in this system. All the lamps, except SHPs, were fed into the fluorescent powder containing lamp crushing machine. The fluorescent powder was then separated and collected with other heavy metals by a fan. The SHPs were feed into the airtight high pressure mercury lamp crushing machine by a transport system then crushed automatically under airtight negative pressure conditions. The heavy metals, electrodes, and mercury-containing cullet were collected separately. Gener-



Fig. 1. Schematic diagram of full-scale thermal desorption process to recover the mercury from high-mercury-containing lamps.



Fig. 2. Bench-scale thermal desorption process.

ally, the sizes of crushed SHP particles varied between 1 and 15 cm, while the sizes of crushed fluorescent tube tailpipes, fluorescent tubes and CCFLs ranged between 0.2 mm and 2 cm.

2.1.2. Thermal desorption system

The thermal desorption system contained a thermal desorption oven and a mercury recovery system, divided into a feed-in unit, thermal desorption oven, condenser, and activated carbon adsorption tank. The overall size of the thermal desorption system was L2990 mm \times W1580 mm \times H2600 mm with a maximum output power of 35 kW and maximum batch thermal desorption treatment time of 24 h. This thermal desorption system needed 10-16 h to treat 1001 of waste lamps. In the feed-in unit, 4 buckets, loaded with 20-301 of mercury-containing lamps per buckets, were hung in the thermal desorption oven. Two heating chambers were used in the thermal desorption oven, i.e., a pre-heating chamber and a post-heating chamber. The pre-heating chamber could operate continuously at 700-800 °C under vacuum conditions to vaporize the mercury from the lamps. It had a batch treatment capacity of about 1201 of mercury-containing lamps. The postheating chamber operated at over 800 °C to ensure that all the mercury from the mercury-containing lamps was vaporized. The mercury vapor gas flowed into the condenser to be condensed into liquid mercury at 25 °C which was collected in a tank. Finally, uncondensed gas was then treated in an activated carbon adsorption tank. Vulcanized activated carbon was used as a sorbent to lower the mercury content enough to meet the Taiwan EPA criteria of 0.3 mg/m^3 in the exhaust gas.

2.2. Operating conditions

The effects of two major parameters, pre-heating temperature and desorption time, on the performance of the full-scale thermal desorption process were investigated to determine the optimal operating conditions. The pre-heating temperatures were set at $350 \,^{\circ}$ C, $450 \,^{\circ}$ C and $525 \,^{\circ}$ C, while the desorption times were 8, 12 and 15 h. In addition, the post-heating and condensing temper-

Thermal desorption efficiency of SHPs

atures were 825 $^{\circ}$ C and 5 $^{\circ}$ C, respectively. The airflow rate of the desorption process was 28001/h.

2.3. Bench-scale thermal desorption batch experiments

In order to find the correct boiling point of different mercury-containing materials we designed a bench-scale thermal desorption process with which to carry out thermal desorption batch experiments. In this bench-scale thermal desorption process a high temperature combustion-heated furnace with 2.5 cm in diameter and a mercury vapor detector were utilized, as shown in Fig. 2. The furnace was heated from 25 °C to 850 °C at a heating rate of 10 °C/min and a flowrate of 1 l/min. The mercury concentration in the exhaust gas was detected every two minutes. Four kinds of samples (i.e., pure mercury, fluorescent tubes, CCFLs contained mercury/fluorescent-powder, and SHPs) were tested at the boiling temperature in this study.

2.4. Analytical methods

The mercury concentration in the lamps was analyzed according to the NIEA M317.01C and NIEA R314.11C procedures specified by the Taiwan EPA (www.niea.gov.tw, in Chinese). Briefly, both untreated and treated-lamps were solubilized in a nitrohydrochloric acid solution. The total mercury concentration was then determined using the PerkinElmer FIMS 400 cold vaporatomic absorption method. In addition, the mercury concentration in the exhaust gas of each experimental batch was analyzed by a mercury vapor analyzer (JEROME 431, Arizona Instruments LLC).

3. Results and discussion

3.1. Thermal desorption performance for SHPs

SHPs were the first mercury-containing lamps that were treated by thermal desorption process in this study. The mercury con-

Thermal desorption time (h)	Pre-heating temperature (°C)	Mercury concentration before treatment (mg/kg)	Mercury concentration after treatment (mg/kg)	Thermal desorption efficiency (%)
	350	15,800	2.94	99.98
8	450	3,400	2.43	99.93
	525	905	0.69	99.92
	350	25,200	2.15	99.99
12	450	4,920	1.11	99.98
12	525	11,100	0.77	99.99
15	350	17,200	1.80	99.99
	450	787	1.53	99.81
	525	2,800	0.67	99.98
Average		9,124	1.57	99.95



Fig. 3. Thermal desorption efficiencies of all conditions in this study.

tent in the SHPs before thermal desorption treatment was in the range of 787-25,200 mg/kg, with an average concentration of 9124 mg/kg, all amounts being higher than the 260 mg/kg limit, as shown in Table 1. Among these SHP samples, most contained more than 2000 mg-mercury/kg, while 4 samples contained more than 10,000 mg-Hg/kg. The amplitude and standard deviation of these SHP samples were 24,413 mg/kg and 8657 mg/kg, respectively. Since SHPs do not contain fluorescent powder, the mercury does not attach evenly to the lamp surface. After thermal desorption treatment, under any operating conditions, the mercury content ranged from 0.67 mg/kg to 2.94 mg/kg, with an average of 1.57 mg/kg, also as shown in Table 1. The amplitude and standard deviation of these treated SHP samples were 2.27 mg/kg and 0.82 mg/kg, respectively. It was also estimated that the thermal desorption efficiency, that is the difference in mercury concentration between the untreated and treated waste samples, was from 99.81% to 99.99%, regardless of the operating conditions, with an average, standard deviation and coefficient of variation of 99.95%, 0.06% and 0.06%, as shown in Fig. 3. This reveals the excellent thermal desorption efficiency of this full-scale process on mercury-containing SHPs. In addition, no significant difference of thermal desorption efficiency was observed when the thermal desorption time was longer than 8 h and the pre-heating temperature was higher than 350°C.

Table 2

Thermal desorptior	n efficiency	of fluorescent	tube	tailpip
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3.2. Thermal desorption performance for fluorescent tube tailpipes

As mentioned above, fluorescent tube tailpipes were also treated by the full-scale thermal desorption process. The results in Table 2 show that before the thermal desorption treatment, the average mercury content in the fluorescent tube tailpipe was 3178 mg/kg, with a range from 541 mg/kg to 12,700 mg/kg, all values being higher than 260 mg/kg. It should be noted that the mercury content in one fluorescent tube tailpipe sample was 4 times higher than the average mercury content. If this sample is excluded, the average mercury content was about 2000 mg/kg, and the amplitude and standard deviation of these SHPs samples were 12,159 mg/kg and 3848 mg/kg, respectively. Table 2 also shows that after the thermal desorption process, the mercury content ranged from 0.01 mg/kg to 1.72 mg/kg, with an average of 0.82 mg/kg. In addition, the amplitude and standard deviation of the treated samples were 1.71 mg/kg and 0.54 mg/kg, respectively. In addition, the thermal desorption efficiencies, under all operating conditions, varied from 99.85% to 99.99%, with the average, standard deviation, and coefficient of variation being 99.95%, 0.05%, and 0.05%, respectively, as shown in Fig. 3. This is also an indication of the excellent thermal desorption efficiency of this full-scale process for fluorescent tube tailpipes, in addition to mercury-containing SHPs. Furthermore, there was also no significant difference in thermal desorption efficiency when the thermal desorption time was longer than 8 h and the pre-heating temperature was higher than 350 °C. This is similar to the results in the SHP treatment experiments.

3.3. Thermal desorption performance for fluorescent tubes containing mercury-fluorescent powder

The third type of mercury-containing lamps treated was fluorescent tubes containing mercury/fluorescent-powder. As shown in Table 3, the average mercury content in the fluorescent tubes before thermal desorption treatment was 1132mg/kg, ranging from 730 mg/kg to 1310 mg/kg. An examination of Table 3 shows that the concentrations were not significantly different. The amplitude and standard deviation of the samples were 581 mg/kg and 234 mg/kg, respectively. However, after thermal desorption treatment, with all operating conditions, the mercury content ranged from 7.29 mg/kg to 38.20 mg/kg, with an average of 19.03 mg/kg, almost 12 and 23 times higher on average than the results obtained for the SHPs and fluorescent tube tailpipes. It can also be seen that the thermal desorption efficiencies (for all operating conditions) were from 97.08% to 99.22%, with the average, standard deviation, and coefficient of variation being 98.30%, 0.74% and 0.75%, respectively, as shown in Fig. 3. This revealed that although the pre-heating tem-

Thermal desorption time (h)	Pre-heating temperature (°C)	Mercury concentration before treatment (mg/kg)	Mercury concentration after treatment (mg/kg)	Thermal desorption efficiency (%)
	350	541	0.65	99.88
8	450	12,700	1.56	99.99
	525	764	0.01	99.99
	350	1,400	0.77	99.95
12	450	3,730	0.58	99.99
-	525	701	1.05	99.85
	350	4,460	1.72	99.96
15	450	3,070	0.45	99.99
	525	1,240	0.57	99.95
Average		3,178	0.82	99.95

Table 3

Thermal desor	ption efficiency	of fluorescent tube	s contained mercu	rv/fluorescent-powder
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Thermal desorption time (h)	Pre-heating temperature (°C)	Mercury concentration before treatment (mg/kg)	Mercury concentration after treatment (mg/kg)	Thermal desorption efficiency (%)
	350	1,310	38.20	97.08
8	450	1,160	19.30	98.34
	525	1,200	16.90	98.59
	350	1,280	23.70	98.15
12	450	730	7.29	99.00
	525	1,300	10.10	99.22
	350	729	20.70	97.16
15	450	1,290	18.00	98.61
	525	1,190	17.10	98.56
Average		1,132	19.03	98.30

Table 4

Thermal desorption efficiency of CCFLs containing mercury-fluorescent powder

Thermal desorption time (h)	Pre-heating temperature (°C)	Mercury concentration before treatment (mg/kg)	Mercury concentration after treatment (mg/kg)	Thermal desorption efficiency (%)
	350	391	111.0	71.61
8	450	541	119.0	78.00
	525	546	36.1	93.39
	350	418	102.0	75.60
12	450	518	74.3	85.66
	525	446	53.8	87.94
	350	444	136.0	69.37
15	450	450	74.3	83.49
	525	495	68.8	86.10
Average		472	86.1	81.24

perature and desorption time did influence the thermal desorption efficiency (for fluorescent tubes containing mercury-fluorescent powder), the maximum difference was only 2.14% among all conditions. In addition, it was also observed that the thermal desorption efficiency slightly increased with increasing pre-heating temperature, regardless of the thermal desorption time. On the other hand, no significant difference in thermal desorption efficiency was observed among different thermal desorption times under the same pre-heating temperature. This indicates that the pre-heating temperature was a more important operating parameter than the thermal desorption time. It was also observed that the highest thermal desorption efficiency of 99.22% was obtained when fluorescent tubes containing mercury-fluorescent powder were treated under a 12 h desorption time and 525 °C pre-heating temperature.

3.4. Thermal desorption performance for mercury-fluorescent powder containing CCFLs

Finally, we treated CCFLs containing mercury/fluorescentpowder. As shown in Table 4, the average mercury content in the mercury-fluorescent powder containing CCFLs before thermal desorption treatment was 472 mg/kg, ranging from 391 mg/kg to 546 mg/kg. These concentrations were much lower than those for the other 3 kinds of mercury-containing lamps treated in this study. It was also observed that the concentrations among the mercury/fluorescent-powder containing CCFLs did not vary significantly. The amplitude and standard deviation of these lamps were 155 mg/kg and 55 mg/kg, respectively, which were less than for the other 3 kinds of lamps. However, unlike the other 3 kinds of lamps, the mercury content was still high after thermal desorption treatment, ranging from 36.1 mg/kg to 136 mg/kg with an average of 86.1 mg/kg, as shown in Table 4. In addition, the average thermal desorption efficiency for the CCFLs was only 81.24%, much lower than for the other 3 kinds of lamps, as shown in Fig. 3. The thermal desorption efficiencies ranged from 69.37% to 93.39%, with average, standard deviation, and coefficient of variation of 81.24%, 8.03% and 9.88%, respectively. With only one set of conditions, that is an 8 h desorption time and 525 °C pre-heating temperature, could we achieve a desorption efficiency of more than 90%. As can also be seen in Fig. 3, the thermal desorption efficiency significantly increased with increasing pre-heating temperature, for all thermal desorption times, indicating that the pre-heating temperature is a more important operating parameter than the thermal desorption time. The optimum conditions for treating CCFLs containing mercury-fluorescent powder were a desorption time of 8 h and a pre-heating temperature of 525 °C.

3.5. Comparison of the thermal desorption efficiencies of the four types of lamps

Fig. 4 shows a comparison of the averaged thermal desorption efficiencies of the types of four lamps used in this study averaged under all conditions. It can be seen that the average thermal des-



Fig. 4. Average thermal desorption efficiencies in this study.



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Fig. 5. Mercury vaporization batch experiments.

orption efficiency of SHPs, fluorescent tube tailpipe, fluorescent tubes containing mercury-fluorescent powder and CCFLs containing mercury-fluorescent powder were 99.95%, 99.95%, 98.30%, and 81.24%, respectively. The differences in thermal desorption efficiency among these four kinds of lamps could be due to the complexity of the mercury compounds in the lamps. For example, the SHPs and fluorescent tube tailpipe contained pure mercury, and the thermal desorption efficiencies of these two types of lamps were both 99.95%. In contrast, the CCFLs, which contained large amounts of mercury-fluorescent powder, had a thermal desorption efficiency of only 81.24%, significantly lower than the thermal desorption efficiency of fluorescent tubes, which contained only a little mercury/fluorescent-powder.

In addition, for the lamps containing only mercury, there was no significant increase in thermal desorption efficiency observed, even when the thermal desorption time exceeded 8h and the pre-heating temperature was higher than 350 °C. This might be because most of the liquid mercury had vaporized into the mercury vapor before 350 °C, as shown in Fig. 5. On the other hand, for the lamps containing complex mercury compounds, such as fluorescent tubes and mercury-fluorescent powder containing CCFLs, a higher pre-heating temperature significantly enhanced the thermal desorption efficiency, perhaps due to the complexity of the mercury compound. Fig. 5 shows the mercury vaporization batch experiments. It can be seen that the mercury in the samples with pure mercury and the SHPs vaporized after 160°C, and that most of the mercury vaporized before the boiling temperature of 357 °C. On the other hand, for the fluorescent tubes containing mercuryfluorescent powder, there was still a lot of mercury to be vaporized after 357 °C, while, for the CCFLs containing mercury-fluorescent powder, most of the mercury vaporized around 680 °C. This can be explained by the results of Raposo et al. [4], who found that different mercury compounds vaporized at different temperatures, and the more complex the mercury compounds, the higher the temperatures at which they vaporized. In Fig. 5 it can be seen that the mercury was completely released in the range from 700 °C to 850 °C, but it is not recommended that a thermal desorption system be operated at such high temperatures due to high energy costs. In addition, in the MRT system, a magnetic separation unit was used to separate the amalgam, glass, fresh and phosphor components from the combined material. It was observed that it was not easy to separate the amalgam, mercury, and phosphor. Unfortunately, the CCFLs contained more complex mercury compounds than the other lamps. Thus, the mercury release behavior was very diverse, as shown in Fig. 5. Furthermore, it was found in our other study (data not shown here) that complete separation the mercury in the CCFLs was released before 200 °C. Consequently, it is suggested that the thermal desorption efficiency can be improved by separating the crushed lamps completely rather increasing the thermal desorption temperature.

4. Conclusion

In this study, we treated SHPs, fluorescent tube tailpipes, mercury-fluorescent powder containing fluorescent tubes, and mercury-fluorescent powder containing CCFLs by a full-scale thermal desorption process, given different pre-heating temperatures and desorption times. It was found that the average mercury contents for SHPs, fluorescent tube tailpipes, mercury-fluorescent powder containing fluorescent tubes, and mercury-fluorescent powder containing CCFLs were 9124 mg/kg, 3178 mg/kg, 1132 mg/kg and 472 mg/kg, respectively. After thermal desorption treatment, the mercury content for the above four samples became 1.57 mg/kg, 0.82 mg/kg, 19.03 mg/kg and 86.1 mg/kg, respectively, which corresponds to removal efficiencies of 99.95%, 99.95%, 98.03%, and 81.24%, respectively. The bench-scale thermal desorption batch experiments showed that the thermal desorption efficiency was influenced by the complexity of the mercury compounds contained in the lamps. For lamps containing mercurycomplex compounds, the thermal desorption efficiency increased with the higher temperatures. On the contrary, for the lamps containing simple-mercury compounds, there was no significant difference in thermal desorption efficiency difference under different thermal desorption temperatures.

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