

Journal of Hazardous Materials B82 (2001) 247-262



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Pollutants in incineration flue gas

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Received 1 July 2000; received in revised form 31 October 2000; accepted 10 November 2000

Abstract

Previous studies have shown that pollutants from incineration include heavy metals, organic compounds, particulate and acid gas. However, most studies on a single pollutant, it is rare for a study to concentrate on all possible pollutants and the relations between these pollutants under various incineration conditions. The objective of this work was to experimentally study the effect of different operating conditions on the pollutants emitted during incineration and the relations between these pollutants. The operating conditions of the experiments included the temperature of the combustion chamber and the species of organics. The findings indicated that the concentration of hydrogen chloride (HCl) in the presence of polyvinyl chloride (PVC) was higher than that of sodium chloride (NaCl). Regardless of what Cl-containing feedstock was added, the concentration of chromium (Cr) was constant. When organic chloride was added, Cr was the main metallic element which influenced the formation of polycyclic aromatic hydrocarbons (PAHs). On the other hand, when inorganic chloride (NaCl) was added, lead (Pb) was the major element. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Incineration; Heavy metal; Acid gas; PAHs; PVC; NaCl

1. Introduction

Incineration is one of the best disposal technologies for municipal solid waste, since it has the advantages of volume reduction and high toxicity reduction. In addition, the heat energy produced during incineration can be recovered for electric power generation. However, inadequate design or operation can result in the emission of pollutants. The pollutants contain heavy metals, acid gases and toxic organic compounds, which pose serious threats to human health. During incineration most heavy metals react with oxygen or chlorine to form metallic oxides or metallic chlorides. Some of the species with low volatility remain

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^{0304-3894/01/\$ –} see front matter © 2001 Elsevier Science B.V. All rights reserved. PII: S0304-3894(00)00355-1

in the sand bed. Conversely, metallic vapors of high volatility, which are carried out with the flue gas, generate condensates during cooling. This process of vapor condensation includes homogeneous nucleation and heterogeneous condensation. The reactive characteristic of heavy metals results from heating during incineration, giving rise to chemical interaction, vaporization, condensation, coagulation and deposition. The chlorides, sulfides and nitrides in municipal solid waste (MSW) react with hydrogen and oxygen to generate hydrogen chloride, sulfuric acid and nitric acid. The generation and control of acid gases is related to the operating temperature, the waste composition and the air pollution control device (APCD) [1-10]. In addition, Wey and Fang [11] indicated that the quantity of heavy metals and the species containing chlorides in MSW could affect the generation of hydrogen chloride. On the other hand, kinetic restrictions result both in incomplete combustion, and also in the formation of trace organics, such as PAHs, dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs). In addition, heavy metals affect the formation of PAHs. Wey et al. [12] found that Cd and Cr in fly-ash provide the activity required for the reaction between PAH precursors and the low-ring PAHs of fly-ash. Cr promoted the formation of five- and six-ring PAHs. Pb provided many more adsorption sites than Cr, but the catalyzing ability of Pb is far lower than that of Cr. The presence of excess O_2 , Fe, Sn and chlorides could catalyze the formation of PCDDs/PCDFs [13,14]. The above studies just focus on single pollutants. The generation of all possible pollutants and the relations between these pollutants under various incineration conditions were rarely studied.

The research presented here, a pilot scale incinerator was used to evaluate the potential pollutants from the incineration of MSW and to understand the relations between these pollutants before the air pollution control device (APCD).

2. Experimental

2.1. Preparation of simulated feed wastes

According to the chemical analytic composition of MSW in Taiwan in 1996 (Table 1), the simulated feed wastes used in the experiments were composed of sawdust, low-density polyethylene (LDPE), PVC (or NaCl), sulfur (S) and a heavy metal solution. Three investigated metals Cd, Pb, Cr (with nitrate) of 0.01, 0.1 and 0.1 wt.%, respectively, were dissolved in the distilled water. Each sample of mixed feedstock described above was enclosed in a polyethylene (PE) bag of 0.25 g. Table 2 lists the detailed composition of various simulated feedstock.

2.2. Apparatus

The reactor for the present study is a bubble fluidized bed incinerator, consisting of a feedstock feeder, a preheated chamber, a primary combustion chamber, and a secondary combustion chamber (Fig. 1). The preheated chamber was 50 cm long. The primary combustion chamber was 110 cm high and 10 cm in diameter. The secondary combustion chamber was 100 cm high and 25 cm in diameter. The incinerator was fitted with a perforated stainless steel gas distributor. Seven thermocouples were used to determine the temperature profile in the preheated chamber, sand bed, freeboard chamber and outlet of freeboard chamber.

Table 1 The chemical analytic composition of MSW in Taiwan in 1996 [17]

Ash (%)	13.79
Combustibles (%)	
С	18.99
Н	2.82
0	13.06
Ν	0.45
S	0.12
Cl	0.16
Heavy metal (%)	
Cd	0.012
Hg	0.00025
Pb	0.1
Cr	0.1
HHV (kcal/kg)	2106.72
LHV (kcal/kg)	1650.69
C/N	41.74

The flue gas was treated by a cooling tower, two consecutive cyclones and a wet scrubber, and then induced into atmosphere.

2.3. Experimental procedure

By calculating the required amount of theoretical air and an excess factor of 60%, 53 l/min input air at room temperature was determined. The effects of sand bed temperature on

Table 2 The composition of feedstock .

Run no.	Feedstock (g per bag)									
	PE ^a bag	LDPE ^b	Sawdust	S	Cd(NO ₃) ₂	Pb(NO ₃) ₂	Cr(NO ₃) ₃	Water	PVC ^c	NaCl
1	0.25	0.12	1.322	0.003	0.00055	0.0042	0.0089	1	0.006	_
2	0.25	0.12	1.322	0.003	0.00055	0.0042	0.0089	1	0.006	_
3	0.25	0.12	1.322	0.003	0.00055	0.0042	0.0089	1	0.006	_
4	0.25	0.12	1.322	0.003	0.00055	0.0042	0.0089	1	_	0.0056
5	0.25	0.12	1.322	0.003	0.00055	0.0042	0.0089	1	_	0.0056
6	0.25	0.12	1.322	0.003	0.00055	0.0042	0.0089	1	_	0.0056
7	0.25	0.12	1.322	0.003	0.00055	0.0042	0.0089	1	0.006	-
8	0.25	0.12	1.322	0.003	0.00055	0.0042	0.0089	1	0.006	_
9	0.25	0.12	1.322	0.003	0.00055	0.0042	0.0089	1	0.006	-
10	0.25	0.12	1.322	0.003	0.00055	0.0042	0.0089	1	_	0.0056
11	0.25	0.12	1.322	0.003	0.00055	0.0042	0.0089	1	_	0.0056
12	0.25	0.12	1.322	0.003	0.00055	0.0042	0.0089	1	-	0.0056

^a PE: polyethylene.

^b LDPE: low-density polyethylene.

^c PVC: polyvinyl chloride.



Fig. 1. The bubble fluidized bed incinerator. (1) blower, (2) flow meter, (3) air preheated chamber, (4) air distributor, (5) primary combustion chamber, (6) secondary combustion chamber, (7) thermo-couples, (8) cooling system for feeding, (9) feeder, (10) burner, (11) cooling tower or spray dryer reactor, (12) cyclones, (13) scrubber, (14) I. D. fan.

formation of various concentration of pollutants were examined by setting the sand bed temperature at 700–900°C. When investigating the effects of freeboard zone temperature on pollutants formation, the sand bed temperature was set at 700°C and the freeboard zone temperature controlled by gas burner (when on or off) was set at 700–900°C. The actual measured temperature and the operating condition of each run were listed in Table 3.

Table 3The operating conditions of each run

Run no.	Temperature (°C	Cl-containing species				
	Primary combust	tion chamber	Average ^a	Secondary combustion chamber		
	Above sand bed	In sand bed				
1	722.2	755	738.6	_	PVC	
2	819.1	853	836	_	PVC	
3	852.4	883.6	868	_	PVC	
4	768	894	831	_	NaCl	
5	837	978	908	_	NaCl	
6	939	1000	970	_	NaCl	
7	725.8	681	703	711	PVC	
8	725	698	712	802	PVC	
9	728	706	717	936	PVC	
10	727	698	713	703	NaCl	
11	729	679	704	807	NaCl	
12	728	664	696	925	NaCl	

^a Average temperature = (above sand bed temperature + in sand bed temperature)/2.

The static sand bed height was 15 cm high and the size of silica sand employed here was about 840 μ m. The corresponding minimum fluidization velocity was 20.92–23.86 cm/s. The chamber was heated by electrical heaters, and when the set temperature reached a steady state, every synthetic feed waste was fed into the incinerator at regular interval of 20 s.

2.4. Methods of sampling and analysis

2.4.1. Metals

The flue gas containing heavy metal pollutants was sampled by a stainless sampling probe and passed through the filter holder contained a glass fiber filter, which collected particles, then passed through impingers to absorb the remaining metals. Isokinetic sampling was performed. The sampling position was located after secondary combustion chamber and the sampling apparatus is shown in Fig. 2, which refers to the standard USEPA method 5 (M5). The impingers were submerged in an ice bath to enhance condensation, and the mixed absorption solution of impingers were 200 ml 5% HNO₃ and 10% H₂O₂. After the sampling train was completed, the metal samples contained glass fiber filter and absorption solution were pretreated by microwave digestion, and the recovery efficiency was measured. The concentration of metals was then analyzed by atomic adsorption spectroscopy (Hitachi Model Z-8000 Polarized Zeeman AAS) according to USEPA method 3050.

2.4.2. HCl, SO_2 and NO_x

The same sampling method for metals was employed to sample HCl as well (Fig. 2). The impingers with 100 ml 0.1N NaOH absorption solution were used to adsorb the remaining



Fig. 2. The sampling train for HCl or heavy metal. (1) sampling probe, (2) thermometer, (3) filter holder, (4) impingers, (5) silica gel, (6) flow meter, (7) connect of vacuum pump.



Fig. 3. The sampling train for PAHs. (1) sampling probe, (2) heated filter and heating hose, (3) thermometer, (4) cooling tube and XAD-4 adsorption tube, (5) impingers, (6) 200 ml. distilled water, (7) silica gel, (8) flow meter, (9) connect of vacuum pump.

HCl. The analysis of HCl was performed by the method of colorimetry using mercuric thiocyanate (ROC EPA NIEA method A412.70A). The concentration of SO₂ and NO_x were detected using a flue gas analyzer (BACHARACH model 300). The analyzer was corrected using the standard gas with determinate concentration before the experiment process. The range of concentration for SO₂ and NO_x monitoring was 0–1999 ppm, and their accuracy was $\pm 5\%$ or ± 10 ppm.

2.4.3. Organic compounds

The same sampling location used for metals was used to sample organic compounds as well. Isokinetic sampling was also performed. The USEPA modified method 5 (MM5) was used for sampling (Fig. 3). The flue gas containing organics was sampled using a stainless sampling probe and passed through the heated filter packed with glass fiber, which removed particles. Then it was passed through a cooling tube and the remaining organics was captured using an adsorption tube packed with 6 g XAD-4 resin. Furthermore, the organic samples were initially extracted for 20 h using a Soxhlet extraction process and the solution was concentrated to 1 ml using a KD evaporative concentrator. Finally, the samples were put into 1.8 ml brown vials and stored at 4°C. These samples were later (in 14 days) analyzed with a GC/FID detector (Perkin-Elmer Autosystem GC). The recovery efficiency and multiple analyses were carried out to identify the precision and accuracy of above analytical procedure. The recovery efficiency of PAHs is about 60.6–99.7%, and the standard deviation of the PAHs analysis is about 1.1–15.3.

3. Results and discussion

3.1. Solid-phase pollutants

Most of the light and small particles emitted with the flue gas during incineration are carbon-based. The concentration and properties of these particles not only are relative to the phenomenon of homogeneous nucleation and heterogeneous condensation but also affect the efficiency of the air pollution control device (APCD). Fig. 4 shows the comparison of particulate concentration under various operating conditions during the incineration, indicating that the operating conditions of the secondary combustion chamber reduce the particulate concentration. Additionally, when the burner was off (only with sand bed combustion, Fig. 4(a)), the particulate concentration increased if inorganic chloride (NaCl) was added. This could be because the sodium is conversed to sodium ion at high temperature and then combines with other atom forming novel species. Therefore, the particulate concentration is higher than the presence of organic chloride (PVC). When the burner was on (with sand bed combustion and freeboard combustion), however, the influence of combustion efficiency was more than the chloride additives (Fig. 4(b)). On the other hand, when the burner



Fig. 4. The concentration of particles under various operating conditions: (a) burner off; (b) burner on (the sand bed temperature was set at 700° C).



Fig. 5. The generation of Pb under various chloride additives: (a) burner off; (b) burner on (the sand bed temperature was set at 700° C).

was off, the solid wastes would not have sufficient residence time for high temperature combustion and, as a result, would produce lots of unburned carbons.

3.2. Heavy metal pollutants

The main objective of operating the burner is to make the products of incomplete combustion (PICs) reach complete combustion. The species, which enter the secondary combustion chamber with the flue gas, are the vapors of heavy metal, metallic compounds of high volatility and traces of bottom ash. These heavy metals and their compounds may convert to other types of metallic compounds by recombining in the secondary combustion chamber. Figs. 5 and 6 present the effects of various chloride additives on Pb and Cd formation. Fig. 5(a) indicates that the concentration of Pb increases when inorganic chloride (NaCl) was added and the burner was off. When the burner was on (Fig. 5(b)), however, the presence of PVC increases the concentration of Pb as well. The results of Fig. 6 are the same as Fig. 5. Wey et al. [2] and Chen et al. [3,4] indicate that when organic chloride (PVC) was added, the potential for metallic chloride formation is higher than that of metallic oxide during incineration process and the distribution of metal in the sand bed would be reduced due to the high volatility of metallic chlorides. Moreover, the possibility



Fig. 6. The generation of Cd under various chloride additives: (a) burner off; (b) burner on (the sand bed temperature was set at 700° C).

of chlorine that reacts with the metal during incineration is small because the affinity of chlorine and sodium is stronger than that of metal. Most metals react with oxygen to form metallic oxides, which have a high boiling point and remain in the sand bed. The concentration of most metals tends to be reduced when inorganic chloride (NaCl) was added. Fig. 7 presents the effect of various chloride additives on Cr emission. It can be observed that the effect of temperature on the emission concentration of Cr is small, regardless of the presence of organic or inorganic chloride. Some researchers [2,15] indicate that the main chromium specie is $Cr_2O_3(s)$ accounting for its stability at the temperature of 600–1200 K during incineration, and a trace of $CrO_2Cl_2(g)$ is generated as the amount of chlorine increases.

3.3. Acid gas pollutants

Fig. 8 presents the effect of various chloride additives on HCl formation, indicating that the concentration of HCl in the presence of organic chloride (PVC) is slight higher than that in the presence of inorganic chloride (NaCl). This is because that PVC can provide HCl with the source of hydrogen, so the chlorine ions can connect with hydrogen ions to form HCl at high temperature rapidly. Nevertheless, NaCl do not provide the source of



Fig. 7. The generation of Cr under various chloride additives: (a) burner off; (b) burner on (the sand bed temperature was set at 700° C).

hydrogen inversely, the formation of HCl must depend on water in the waste. Moreover, the affinity between sodium and hydrogen is larger than chlorine and hydrogen. Therefore, the concentration of HCl in the presence of inorganic chloride (NaCl) is slight lower than that in the presence of organic chloride.

Fig. 9 presents the effect of various chloride additives on SO_2 formation, indicating that the concentration of SO_2 decreases with the temperature increases in the presence of inorganic chloride. However, when organic chloride was added, the concentration of SO_2 has no significant changes.

Fig. 10 presents the effect of various chloride additives on NO_x formation during the incineration process. The results indicate that the chloride additives do not influence the concentration of NO_x regularly. Comparing Fig. 10(a) and (b), when the burner was on, the concentration of NO_x was higher than when the burner was off. The main reason for this phenomenon is that thermo- NO_x or fuel- NO_x are produced at higher temperature. Although the average temperature of primary combustion chamber (burner off) was similar to the temperature of secondary combustion chamber (burner on), the local high temperature generated by burner would result in the formation of thermo- NO_x . In addition, the fuel of burner employed in the present study is the liquefied natural gas that contains nitride. Therefore, the fuel- NO_x is possibly produced during incineration as well.



Fig. 8. The generation of HCl under various chloride additives: (a) burner off; (b) burner on (the sand bed temperature was set at 700° C).



Fig. 9. The generation of SO_2 under various chloride additives.



Fig. 10. The generation of NO_x under various chloride additives: (a) burner off; (b) burner on (the sand bed temperature was set at 700°C).

3.4. Organic pollutants

Table 4 lists the classifications and abbreviation of organic compounds studied at present study. Fig. 11 presents the effect of temperature on the PAHs formation in the presence of PVC when the burner was off, indicating that the main species of PAHs are naphthalene (NaP) and acenaphthylene (AcPy) when the temperature of the primary combustion chamber reaches 738°C. But these compounds are gradually converted to fluorine (Flu), phenanthrene (Pha) and anthracene (Ant) as the temperature increases. The main result is that the electronegative charge of chlorine is strong, and it may connect to the alkene or alkyne, and the organic would recombine to form stable ring-species.

Fig. 12 presents the effect of temperature on the PAHs formation in the presence of inorganic chloride (NaCl) when the burner was off, indicating a wide range of organic species is generated. However, most organic compounds are destroyed at higher temperature (970°C) to form CO_2 and H_2O . It indicates that the concentrations of organic compounds decrease with increase in temperature.

Table 4 Classifications and abbreviation of organics studied at present study

Multi-ring compounds (PAHs)	Abbreviation
Two-ring compounds	
Naphthalene	Nap
Three-ring compounds	
Acenaphthylene	AcPy
Acenaphthene	AcP
Fluorene	Flu
Phenanthrene	Pha
Anthracene	Ant
Four-ring compounds	
Fluoranthene	FluA
Pyrene	Pyr
Benzo(a)anthracene	B(a)A
Chrysene	Chr
Five-ring compounds	
Benzo(b)fluoranthene	B(b)F
Benzo(k)fluoranthene	B(k)F
Six-ring compounds	
Benzo(a)pyrene	B(a)P
Dibenzo(a,h)anthracene	DbA
Benzo(ghi)perylene	B(ghi)P
Indeno(1,2,3-cd)pyrene	InP



Fig. 11. The effect of temperature on PAHs formation (PVC feed; burner off).



Fig. 12. The effect of temperature on PAHs formation (NaCl feed; burner off).

Fig. 13 presents a comparison of the concentration of PAHs in the presence of organic chloride when the burner was on or off. The results show that most PAHs were destroyed at higher temperature (burner on). Only a few PAHs were converted to other high-ring PAHs after the bonds were broken. The organic compounds gradually convert to three-, fourand even five-ringed as the temperature increases. When the temperature of secondary combustion chamber is 711°C, the main organic pollutants was two- or three-ring PAHs.

Fig. 14 presents a comparison of the concentration of the PAHs in the presence of inorganic chloride (NaCl) whether the burner was on or off. The results reveal that when



Fig. 13. The comparison of the concentration of PAHs in the presence of PVC whether the burner was on or off.



Fig. 14. The comparison of the concentration of PAHs in the presence of NaCl whether the burner was on or off.

the burner was off, most PAHs species had a three- and four-ring structure. Nevertheless, when the burner was on, the higher temperature could effectively decompose three- and four-ring PAHs and then a portion of them would recombine forming PAHs of more rings at higher temperature. Therefore, the concentrations of high-ring structure of PAHs increase slightly.

Wey et al. [16] indicated that the presence of Pb can reduce the activation energy required for the reaction between precursors and low-ring PAHs in the gas phase, so the concentration of fluoranthene (FluA), pyrene (Pyr), benzo(a)anthracene (B(a)A), chrysene (Chr) and benzo(a)pyrene (B(a)P) are higher than that of other species at high temperature. Cd and Cr in the fly-ash provided the activity required by the reaction between PAHs precursors and the low-ring PAHs of fly-ash. Cr prompted the formation of five- and six-ring PAHs, for example, the concentration of PhA, FluA, Pyr, indeno(1,2,3-cd)pyrene (InP) and benzo(ghi)perylene (B(ghi)P) are higher than that of other species. In addition, Pb provides many more adsorption sites than Cr, although the catalyzing ability of Pb is far lower than that of Cr.

The concentrations of PhA, FluA, Pyr and B(a)A in Fig. 13 are higher than those of other species. The concentrations of B(a)A, Chr and B(a)P in Fig. 13 are clearly higher than the others. It could be concluded that the situation in Fig. 13 is due to the influence of Cr, and the presence of Pb influences the results of Fig. 14. Compared to Figs. 5 and 7, the concentration of Pb in the presence of PVC is lower than that in the presence of NaCl, although the concentration of Cr is opposite. So the main result for this phenomenon in Fig. 14 is the catalyzing ability of Cr in the presence of PVC is far higher than that of Pb. When inorganic chloride was added, although the catalyzing ability of Cr is higher than that of Cr. The effect of catalysis of Pb on PAHs is more significant.

4. Conclusions

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The main objective of the present study was to study the pollutants emitted from the incineration of MSW and the relations between these pollutants under various operating conditions. The results show that the concentration of Cd increased as the temperature increased. The concentration of Pb in the presence of inorganic chloride (NaCl) increased as the temperature increased. Regardless of what Cl-containing feedstock was added, the concentration of Cr was constant. In addition, the concentration of HCl in the presence of PVC was higher than that of NaCl. When organic chloride was added, the PAH species, which were not destroyed at high temperature would convert from a two- to a three-ring structure PAHs as the temperature increased. When organic chloride was added, Cr was the main metallic element which influenced the formation of PAHs. On the other hand, when inorganic chloride (NaCl) was added, Pb was the major element.

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